SERPENTINE AND ASSOCIATED ROCKS AND CONTACT MINERALS NEAR WESTFIELD, MASSACHUSETTS

by

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Serpentine and Associated Rocks and Contact Minerals Near Westfield, Massachusetts

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Submitted to the Department of Geology & Geophysics
on January 11, 1960 in partial fulfillment of the
requirements for the degree of Ph.D.

Serpentine occurs at Westfield, Massachusetts associated with amphibolite and impure marble in several thin lenses intercalated in steeply dipping pelitic schist. The lenses are restricted to a thin zone about 1000 feet wide and exhibit nearly complete textural and mineralogical gradations between the three rock types. They vary in thickness from a few inches to five hundred feet and in length from six inches to 3/4 mile. The schist and lens rocks are intimately intruded by quartz diorite. Where the quartz diorite has intruded serpentine or marble, thin monomineralic bands of biotite and actinolite have developed parallel to the contact, with the biotite band next to the intrusive.

Twelve biotite and seven actinolite samples from one area of the monomineralic banding have been analyzed in duplicate by rapid analysis methods. The standards G-1, W-1, and Haplogranite were analyzed at the same time and under the same conditions. The ranges of variation of the major element concentrations in the biotites are SiO_2: 37.93 - 42.05%; Al_2O_3: 14.26 - 19.22%; FeO: 2.90 - 7.96%; Fe_2O_3: 1.13 - 3.04%; MgO: 17.98 - 24.42%; and K_2O: 8.96 - 10.31%. In the actinolites the ranges are SiO_2: 51.33 - 58.34%; Al_2O_3: .02 - 7.70%; FeO: 2.28 - 3.90%; Fe_2O_3: .56 - 1.74%; MgO: 19.49 - 24.90%; and CaO: 10.04 - 13.34%.

Results from analyses of biotite-actinolite pairs taken at varying distances from the contact between the two bands indicate culminations and depressions perpendicular to the contact in the content of the various elements of the two minerals. The variation in the Mg/Fe ratios in the two minerals indicates a degree of chemical disequilibrium in the bands, since it can be shown that attainment of equilibrium requires that the Mg/Fe ratios in different samples of the same mineral be similar.

The mineralogy and replacement relationships of the lens rocks are best explained by regional metamorphism of dolomite rock with varying amounts and types of original impurity. The monomineralic banding is viewed strictly as the result of a contact reaction between the intrusive quartz diorite and the serpentine-marble.

Thesis Supervisor: H. W. Fairbairn
Title: Professor of Geology

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INTRODUCTION

The general thesis topic and specific problems studied were chosen to further the author's field experience, and at the same time to make use of the rapid analysis laboratory in the Department of Geology and Geophysics at MIT. The author participated in the organization and initial use of this laboratory, which was part of a National Science Foundation project directed by Professors H. W. Fairbairn and Gordon J. F. MacDonald. The general aim of this project was to investigate the extent to which chemical equilibrium has been attained in metamorphic processes. The chemical work reported here is along these lines, dealing with the analysis of co-existing minerals. The development of rapid analysis techniques will allow detailed chemical studies on a larger scale than has been possible in the past. Such studies will in the future provide an important means of studying many of the problems of igneous and metamorphic geology. The theoretical discussion indicates how chemical analyses might be used for quantitative applications of thermodynamics to geologic problems. Finally, the general geology and petrography resulting from the field work represent a small contribution to the geology of serpentine rocks.

Part Two is a presentation of the entire thesis. Part One is a condensation of Part Two, prepared in the form of a manuscript ready for publication.

PART I

Abstract

Serpentine occurs at Westfield, Massachusetts associated with amphibolite and impure marble in several thin lenses intercalated in steeply dipping pelitic schist. The lenses are restricted to a thin zone about 1000 feet wide and exhibit nearly complete textural and mineralogical gradations between the three rock types. They vary in thickness from a few inches to five hundred feet and in length from six inches to 3/4 mile. The schist and lens rocks are intimately intruded by quartz diorite. Where the quartz diorite has intruded serpentine or marble, thin monomineralic bands of biotite and actinolite have developed parallel to the contact, with the biotite band next to the intrusive.

Twelve biotite and seven actinolite samples from one area of the monomineralic banding have been analyzed in duplicate by rapid analysis methods. The standards G-1, W-1, and Haplogranite were analyzed at the same time and under the same conditions. The ranges of variation of the major element concentrations in the biotites are SiO_2: 37.93 - 42.05%; Al_2O_3: 14.26 - 19.22%; FeO: 2.90 - 7.96%; Fe_2O_3: 1.13 - 3.04%; MgO: 17.98 - 24.42%; and K_2O: 8.96 - 10.31%. In the actinolites the ranges are SiO_2: 51.33 - 58.34%; Al_2O_3: .02 - 7.70%; FeO: 2.28 - 3.90%; Fe_2O_3: .56 - 1.74%; MgO: 19.49 - 24.90%; and CaO: 10.04 - 13.34%.

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The mineralogy and replacement relationships of the lens rocks are best explained by regional metamorphism of dolomite rock with varying amounts and types of original impurity. The monomineralic banding is viewed strictly as the result of a contact reaction between the intrusive quartz diorite and the serpentine-marble.

INTRODUCTION

Purpose

Petrographic studies of mineral and rock associations found at serpentine and marble contacts have often been carried out. Bulk chemical analyses have likewise been made in many instances. Little is known, however, about the variation in composition of co-existing contact minerals. This study provides a quantitative idea of the amount of mineral compositional variation in monomineralic biotite and actinolite bands found at the contact of serpentine-marble with intrusive quartz diorite. Among the samples analyzed are five pairs of co-existing biotite-actinolite, taken at different distances from the contact between the two bands. The analyses of these pairs gives an indication of the direction of movement of the various elements and of the relation of the composition of the minerals to distance from the contact. Utilizing the chemical results a method is presented here for testing the degree of attainment of thermodynamic equilibrium in such bands. This work is also an example of the precision and accuracy of rapid analysis procedures as applied to minerals, since all samples were analyzed in duplicate and the standards G-1, W-1, and Haplogranite were run at the same time and under the same conditions. Finally the field and petrographic work represents a small contribution to the problems of serpentine rocks.

Previous Work

Monomineralic banding at the contacts of serpentine bodies is well known. Phillips and Hess (1936) discuss contact

effects between serpentine and siliceous country rocks as shown in the northern Appalachians. Two main types are distinguished: 1) the low-temperature type is marked by formation of a band of chlorite at the edge of the country rock schist and of talc at the edge of the serpentine; 2) the high-temperature type has a band of biotite at the edge of the country rock and of actinolite at the edge of the serpentine. They believe that these effects are caused by hydrothermal solutions which allowed migration of material across the contacts, together with some introduction of material. Read (1934) deals with small round bodies of rock from the Shetland Islands which are made up of circular monomineralic zones. The zoning tends towards the following ideal sequence from the center outwards: antigorite, talc, actinolite, chlorite, biotite, country rock. Read suggests that the zoned masses originally consisted of periodotitic rock, with movement of material during metamorphism producing the zoning. He believes that these rocks cannot be explained on the Appalachain basis of successive formation of minerals with falling temperature and suggests that simultaneous formation has occurred here. Similar zoning in nodules of serpentine in California have been discussed by MacDonald (1941) and Pabst (1942). Other authors who describe such banding include Chidester et. al. (1951), DuRietz (1935), Gibson (1927), and Haapla (1936).

Only seven analyses of contact specimens of biotite and one of actinolite could be found in the literature (Table 3). Relatively little work has been published on the variation in

chemical composition of tremolite-actinolite, although biotite has received as much attention as any mineral. Hall (1941) discusses the relation between color and composition in biotites, and presents fifty-six analyses of biotites for MgO, FeO, TiO_2, and Fe_2O_3, mostly taken from the literature up to 1941. Heinrich (1946) gives seventeen analyses of biotites representing the maximum content of the various constituents which may occur in the mineral. A proposed relation between chemical composition and paragensis in biotites of igneous rocks is outlined by Nockolds (1947). He gives a total of seventy-seven biotite analyses. A broad study of all the micas can be found in Heinrich et al (1953).

GENERAL GEOLOGY

Area Mapped

During 1957 and 1958 an area of about 2 square miles was mapped in detail in the serpentine belt of western Massachusetts. The area consists of two abandoned serpentine quarries and the immediately surrounding rocks and is on the western edge of the Connecticut Valley, about eleven miles west of Springfield and about four miles west of Westfield. The serpentine occurrences are part of the Massachusetts section of the eastern North America serpentine belt (Figure 1). A descriptive study of the serpentine area can be found in Emerson (1898).

Field Relations

Most of the area is made up of pelitic schist and coarse-grained quartz diorite. The schist has the texture and mineralogy

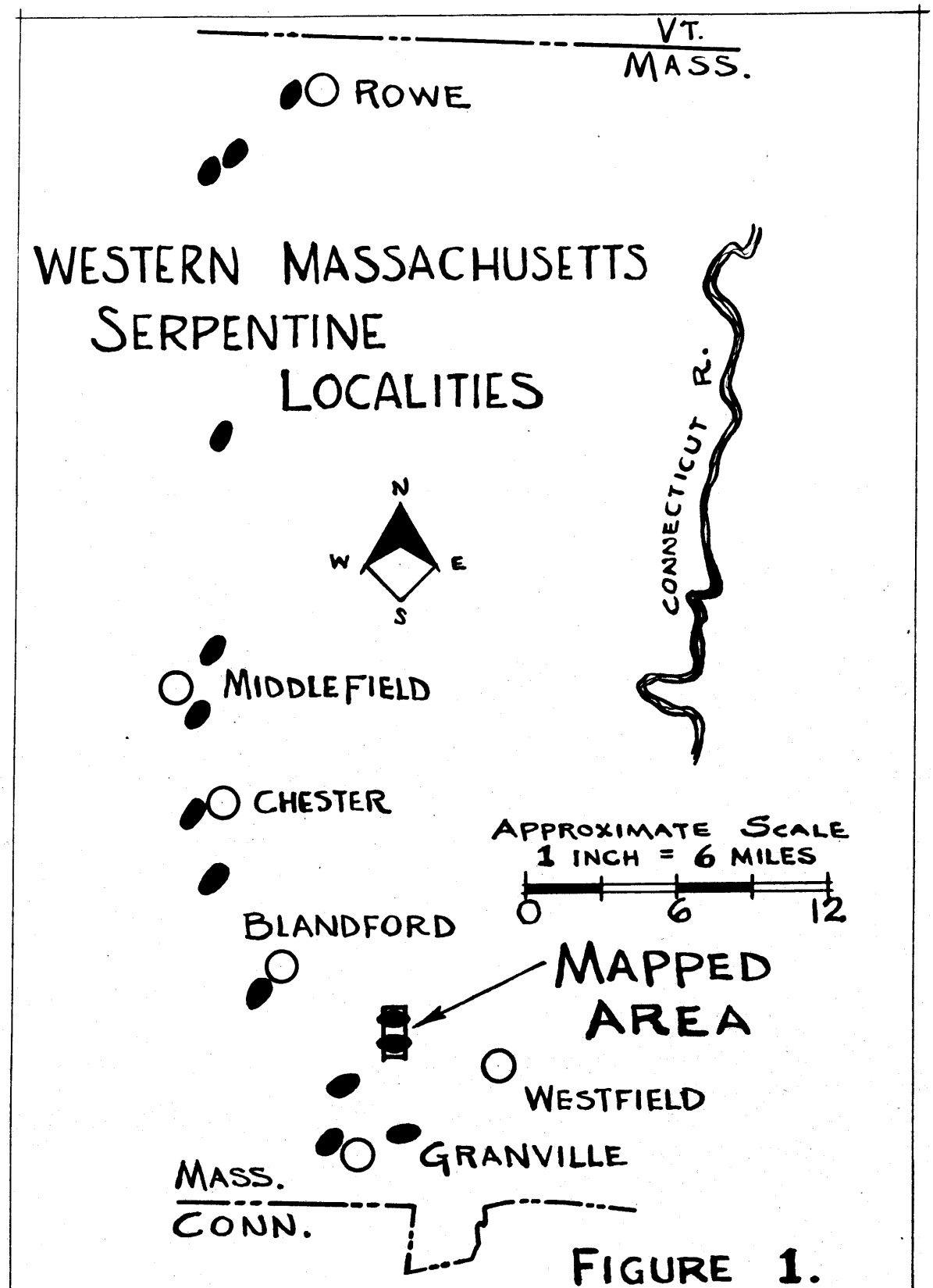


FIGURE 1.

AFTER EMERSON 1898

of a typical high-grade pelitic schist. Porphyroblasts of kyanite and garnet occur in a fine-grained schistose ground-mass of biotite, muscovite, oligoclase, quartz, and magnetite. The dip of the schist varies from vertical to 60° east or west, and is gently warped, varying in strike from N25E to N25W. Everywhere quartz diorite intrudes the schist as large separate masses and as small intimate veins and small masses which appear to grade into the schist. Throughout the area the quartz diorite apparently consists only of quartz, muscovite, and oligoclase, with rare apatite or beryl.

Along a north-south line through the quarries several thin lenses composed of varying combinations of serpentine, amphibolite, and impure marble are intercalated in the schist. These vary in thickness from a few inches to five hundred feet and in length from six inches to 3/4 mile. They also are intruded by quartz diorite. The amphibolite and marble are banded and somewhat schistose. The serpentine is sometimes banded and schistose. The strike of the lenses agrees with that of the enclosing schist, which wraps around the ends of the larger lenses.

The lenses, which are restricted to a thin zone about 1000 feet wide, in places exhibit nearly complete textural and mineralogical gradations between amphibolite, serpentine, and impure marble. In the vicinity of the northern quarry the amphibolite varies from a banded quartz-actinolitic hornblende-andesine rock through chlorite- and epidote-bearing rocks to massive serpentine with lesser carbonate and talc. At the southern

quarry banded actinolite marble passes directly into banded serpentine marble and then into massive serpentine. Thin sections from these rocks exhibit all degrees of replacement of tremolite-actinolite by antigorite, often in association with carbonate. In addition, medium-grained to coarse-grained masses of forsterite show strong replacement by these minerals. Diopside-actinolitic hornblende-epidote-calcite-biotite rock is found south of the southern quarry. Elsewhere smaller lenses show various gradations between the quartz-bearing amphibolite and actinolitic hornblende-epidote-andesine amphibolite. Details of the field and petrographic work are available elsewhere (Brownlow, 1960).

Monomineralic Banding

Where the quartz diorite has intruded serpentine or marble, thin monomineralic bands of mica and actinolite have developed at the contact. These vary from less than an inch to one foot in width, with the actinolite next to the marble or serpentine and the biotite next to the quartz diorite. All contacts are generally sharp. Either or both of the contact minerals may grow perpendicular to a particular contact. At one small area at the southern quarry similar banding is found between schist and serpentine-marble. No other contact effects are visible in the marble, but talc has often developed in the serpentine. In places abundant black tourmaline occurs in the quartz diorite next to the biotite band. Adjacent to the biotite band the quartz diorite is more or less fine-grained, but grades rapidly to its normal coarse grain within a few inches to a foot. Also, near the contact the

quartz diorite has noticeably less quartz and sometimes carries biotite in place of, or with, muscovite.

CHEMICAL ANALYSES

Preparation of Samples

Twelve biotite and seven actinolite samples from adjacent bands of these minerals at the contact between quartz diorite and serpentine-marble have been analyzed in duplicate by rapid analysis methods. The location of these samples, which are all from the southern quarry, is shown in Figure 2. Unfortunately, owing to a lack of adequate fresh samples in the field, the sampling is of a somewhat random nature. Ideally several series of samples in each band perpendicular to the contact should be analyzed, since it is likely that a variation in chemical composition of the two minerals occurs perpendicular to the contact.

From each field sample a rough cube, approximately 1" on a side, was cut. The cube was then broken up by mortar and pestle. That part of the sample which passed through a 60 mesh screen, but not through a 140 mesh screen, was kept. Next the sample was purified to 99% or better by as many as twenty passes through a Frantz Isodynamic Magnetic Separator. Purity and identity of impurities were checked by sample immersion in refractive index liquids. Minerals other than biotite and actinolite found in one or more samples were apatite, talc, magnetite, muscovite, tourmaline, and andesine.

All of the actinolite samples had the same beta index ($1.630 \pm .003$), as determined in white light. Similarly, all the biotite samples, with the exception of #172, had a beta=gamma

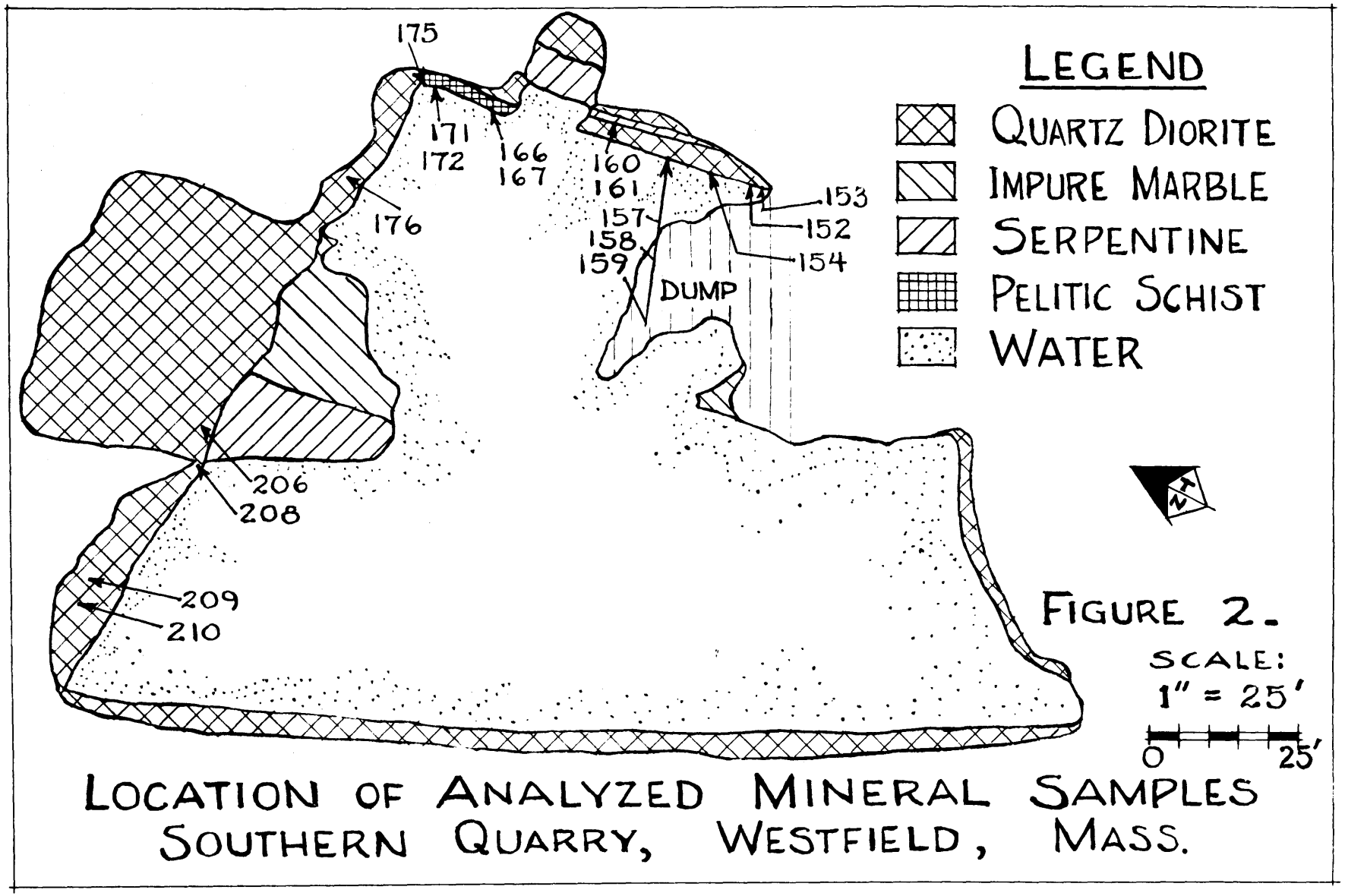


FIGURE 2.

LOCATION OF ANALYZED MINERAL SAMPLES
SOUTHERN QUARRY, WESTFIELD, MASS.

index of 1.597 ± .003. For #172, beta=gamma = 1.608 ± .003. The actinolite was colorless to very light green, and slightly pleochroic from light green to light blue-green. The biotite was found to be very light grey-green and non-pleochroic, again with the exception of #172, which was light to dark brown and non-pleochroic.

Procedures

The analytical procedures which were used are based on the work of Shapiro and Brannock (1956). A somewhat different scheme of analysis has been published by Riley (1958). SiO_2, Al_2O_3, Fe_2O_3, TiO_2, P_2O_5, and MnO were determined by measuring with a spectrophotometer the transmission of light through sample solutions. An automatic photometric titration technique, using a spectrophotometer and recorder, was used for CaO and MgO. K_2O and Na_2O were measured on a Perkin-Elmer Model 146 flame photometer using lithium as an internal standard. Ferrous iron was found by the conventional titrimetric method. Samples weighing .85 grams were used in a single analysis for the above elements, with .40 grams of this amount used for the ferrous iron determination.

In order to drive off all of the water from minerals such as micas and amphiboles it is necessary to have special equipment, since temperatures of the order of 1200°C must be reached. Such equipment was not available and the water content of the minerals was not determined. This is, of course, unfortunate, because the standard chemical check of addition to 100% is eliminated. However, numerous analyses of garnets carried out by

these methods did give an excellent 100% check (W. C. Phinney, personal communication). A calculation of mineral formulas from the analyses given here is still possible, and the results of such calculations may be found in Table I. The calculations were made by assuming the difference between the total of the chemical work and 100% to be all water, and using an atomic basis of 12 (O, OH) atoms for biotite and 24 (O, OH) atoms for actinolite. The water content of G-1 and W-1 was determined using the modified Penfield method of Shapiro and Brannock.

The results of the analyses of the three standards and of the biotite and actinolite samples are given in Tables 1 and 2. Table 3 shows previously published analyses of seven contact biotite samples and one contact actinolite sample. The biotite and actinolite samples analyzed by Phillips and Hess (1936) were taken at the contact between serpentine and schist at Chester, Vermont, and have also been calculated to an atomic basis to provide a comparison with the work reported here. To provide another comparison, two commercial laboratories analyzed biotite #210. The results reported by these laboratories are included in Table 2.

A THERMODYNAMIC TEST OF MINERAL EQUILIBRIUM

If the monomineralic banding has formed strictly as the result of local interaction between two chemically different rock types rather than by introduction of new material by hydrothermal solutions, then necessary conditions of the equilibrium state to which this process would tend can be outlined. Although the possible attainment of these conditions cannot be tested with respect

Table 1. Chemical Analyses and Atomic Calculations
((Biotite Basis: 12 (O,OH) Atoms))

Sample	Biotite #152		Ave.	Biotite #153		Ave.	Biotite #157		Ave.
SiO_2 %	38.74	39.40	39.07	38.05	38.00	38.03	39.30	39.28	39.29
atoms			2.758			2.704			2.780
TiO_2 %	.10	.10	.10	.12	.13	.13	.04	.02	.03
atoms			.005			.007			.002
Al_2O_3 %	15.89	15.81	15.85	17.58	18.05	17.81	18.21	18.39	18.30
atoms			1.318			1.492			1.526
Fe_2O_3 %	1.89	1.81	1.85	1.84	1.98	1.91	1.99	1.83	1.91
atoms			.098			.102			.102
FeO %	5.64	5.60	5.62	7.78	7.86	7.82	5.00	5.04	5.02
atoms			.332			.465			.297
MnO %	.05	.08	.07	.10	.10	.10	.02	.04	.03
atoms			.004			.006			.002
MgO %	20.29	20.05	20.17	18.15	18.03	18.09	20.75	21.10	20.93
atoms			2.121			1.916			2.206
CaO %	1.54	1.60	1.57	.62	.72	.67	none	none	none
atoms			.119			.051			none
Na_2O %	.20	.30	.25	.23	.21	.22	.25	.18	.22
atoms			.034			.030			.030
K_2O %	9.26	8.86	9.06	9.22	9.35	9.29	10.08	9.62	9.85
atoms			.815			.842			.888
P_2O_5 %	1.45	1.49	1.47	.69	.67	.68	.11	.09	.10
atoms			.088			.041			.006
H_2O %	not determined			not determined			not determined		
atoms			2.316			2.489			2.038
Total %	95.05	95.10	95.08	94.38	95.10	94.75	95.75	95.59	95.68

Table 1. Chemical Analyses and Atomic Calculations
((Biotite Basis: 12(O,OH) Atoms))

Sample	Biotite #158		Ave.	Biotite #160		Ave.	Biotite #167		Ave.
SiO_2 %	40.31	40.14	40.22	42.14	41.97	42.05	40.14	39.83	39.98
atoms			2.846			2.955			2.836
TiO_2 %	.16	.14	.15	.02	.06	.04	.76	.74	.75
atoms			.008			..002			.040
Al_2O_3 %	15.69	16.21	15.95	14.09	14.44	14.26	15.82	16.23	16.02
atoms			1.330			1.181			1.345
Fe_2O_3 %	1.57	1.59	1.58	1.59	1.61	1.60	1.96	2.08	2.02
atoms			.084			.084			.108
FeO %	5.72	5.62	5.67	4.95	5.05	5.00	5.99	5.98	5.99
atoms			.335			.294			.355
MnO %	.02	.05	.04	.06	.05	.06	.02	.07	.04
atoms			.002			.004			.002
MgO %	21.49	20.92	21.20	22.69	21.99	22.34	21.37	21.64	21.51
atoms			2.235			2.338			2.273
CaO %	none	none	none	none	none	none	none	none	none
atoms			none			none			none
Na_2O %	.25	.30	.28	.22	.19	.21	.35	.32	.34
atoms			.038			.029			.047
K_2O %	9.84	10.17	10.01	9.55	9.93	9.74	9.01	8.91	8.96
atoms			.904			.874			.810
P_2O_5 %	.14	.10	.12	.03	.07	.05	.04	.03	.04
atoms			.007			.003			.002
H_2O %	not determined			not determined			not determined		
atoms			2.255			2.178			2.057
Total %	95.19	95.24	95.22	95.34	95.36	95.35	95.45	95.83	95.65

Table 1. Chemical Analyses and Atomic Calculations
((Biotite Basis: 12 (O,OH) Atoms))

Sample	Biotite #172		Ave.	Biotite #176		Ave.	Biotite #206		Ave.
SiO_2 %	40.42	39.79	40.10	38.05	37.81	37.93	40.14	39.53	39.83
atoms			2.871			2.684			2.818
TiO_2 %	1.13	1.14	1.14	.05	.08	.07	.20	.19	.20
atoms			.061			.004			.011
Al_2O_3 %	16.67	17.15	16.91	19.16	19.28	19.22	16.61	17.06	16.83
atoms			1.427			1.603			1.403
Fe_2O_3 %	2.25	2.43	2.34	3.07	3.00	3.04	1.72	1.89	1.80
atoms			.126			.162			.096
FeO %	7.96	7.96	7.96	4.02	4.11	4.07	5.39	5.28	5.34
atoms			.475			.240			.316
MnO %	.08	.11	.09	.04	.04	.04	.27	.27	.27
atoms			.005			.002			.016
MgO %	17.79	18.17	17.98	21.51	21.78	21.65	20.44	20.57	20.51
atoms			1.917			2.282			2.163
CaO %	none	none	none	none	none	none	.22	.28	.25
atoms			none			none			.019
Na_2O %	.51	.42	.46	.34	.27	.31	.23	.18	.21
atoms			.064			.043			.029
K_2O %	9.03	9.00	9.02	9.44	9.33	9.39	9.96	9.89	9.92
atoms			.823			.847			.895
P_2O_5 %	.04	.03	.04	.06	.05	.05	.37	.34	.36
atoms			.002			.003			.022
H_2O %	not determined			not determined			not determined		
			1.890			1.996			2.114
Total %	95.88	96.20	96.04	95.74	95.75	95.77	95.55	95.48	95.52

Table 1. Chemical Analyses and Atomic Calculations
((Biotite Basis: 12 (O,OH) Atoms))

Sample	Biotite #208		Ave.	Biotite #209		Ave.	Biotite #210		Ave.
SiO_2 %	40.63	40.14	40.39	40.73	40.14	40.44	40.56	39.90	40.23
atoms			2.831			2.821			2.827
TiO_2 %	.11	.10	.11	.12	.11	.12	.05	.04	.05
atoms			.006			.006			.003
Al_2O_3 %	18.29	18.21	18.25	16.06	16.01	16.04	18.65	19.14	18.90
atoms			1.507			1.319			1.565
Fe_2O_3 %	1.15	1.11	1.13	1.49	1.47	1.48	1.29	1.40	1.35
atoms			.060			.078			.071
FeO %	3.45	3.52	3.49	2.90	2.89	2.90	3.40	3.33	3.37
atoms			.204			.169			.198
MnO %	.19	.21	.20	.06	.06	.06	.08	.09	.09
atoms			.012			.004			.005
MgO %	21.15	21.59	21.37	24.27	24.56	24.42	21.17	21.75	21.46
atoms			2.231			2.538			2.246
CaO %	none	none	none	none	none	none	none	none	none
atoms			none			none			none
Na_2O %	.26	.19	.23	.24	.18	.21	.28	.28	.28
atoms			.031			.028			.038
K_2O %	10.34	10.29	10.31	9.57	9.56	9.56	10.24	10.06	10.15
atoms			.922			.850			.909
P_2O_5 %	.13	.11	.12	.11	.08	.10	.09	.08	.09
atoms			.007			.006			.005
H_2O %	not determined			not determined			not determined		
atoms			2.056			2.172			1.888
Total %	95.70	95.47	95.60	95.55	95.06	95.33	95.81	96.07	95.97

Table 1. Chemical Analyses and Atomic Calculations
((Actinolite Basis: 24 (O,OH) Atoms))

Sample	Actin. #152		Ave.	Actin. #154		Ave.	Actin. #159		Ave.
SiO_2 %	53.77	53.36	53.56	51.29	51.36	51.33	56.31	55.96	56.14
atoms			7.571			7.205			7.712
TiO_2 %	.02	.02	.02	.02	.01	.02	.01	none	.01
atoms			.002			.002			.001
Al_2O_3 %	5.78	5.41	5.59	7.90	7.50	7.70	2.07	1.84	1.95
atoms			.930			1.273			.316
Fe_2O_3 %	1.78	1.61	1.70	1.68	1.81	1.74	.73	.87	.80
atoms			.181			.184			.083
FeO %	3.76	3.98	3.87	4.01	3.80	3.90	2.96	3.02	2.99
atoms			.457			.457			.344
MnO %	.26	.25	.26	.18	.19	.19	.12	.09	.11
atoms			.031			.023			.013
MgO %	19.72	19.97	19.85	19.46	19.51	19.49	21.92	22.13	22.02
atoms			4.180			4.075			4.506
CaO %	12.02	12.34	12.18	11.69	12.06	11.88	12.78	12.94	12.86
atoms			1.844			1.786			1.892
Na_2O %	.86	.80	.83	1.06	1.08	1.07	.45	.39	.42
atoms			.227			.291			.112
K_2O %	.45	.44	.45	.84	.87	.86	.23	.27	.25
atoms			.081			.154			.044
P_2O_5 %	.04	.04	.04	.04	.04	.04	trace	.02	.01
atoms			.005			.005			.001
H_2O %	not determined			not determined			not determined		
atoms			1.555			1.666			2.235
Total %	98.46	98.22	98.35	98.17	98.23	98.22	97.58	97.53	97.56

Table 1. Chemical Analyses and Atomic Calculations
((Actinolite Basis: 24 (O,OH) Atoms))

Sample	Actin. #161		Ave.	Actin. #166		Ave.	Actin. #171		Ave.
SiO_2 %	54.02	53.96	53.99	57.09	56.52	56.80	56.57	56.07	56.32
atoms			7.173			7.802			7.755
TiO_2	.01	none	.01	.02	.01	.02	.01	none	.01
atoms			.001			.002			.001
Al_2O_3 %	1.35	1.25	1.30	1.35	1.27	1.31	1.82	1.75	1.78
atoms			.204			.212			.288
Fe_2O_3	.78	.73	.76	.95	.82	.89	.92	1.04	.98
atoms			.076			.092			.101
FeO %	2.84	2.88	2.86	3.42	3.55	3.48	3.82	3.78	3.80
atoms			.318			.399			.437
MnO %	.15	.15	.15	.17	.14	.16	.18	.16	.17
atoms			.017			.019			.020
MgO %	20.59	20.59	20.59	22.04	21.90	21.97	21.77	21.93	21.85
atoms			4.075			4.495			4.482
CaO %	13.14	13.54	13.34	12.03	12.42	12.22	11.78	11.98	11.88
atoms			1.898			1.798			1.752
Na_2O %	.31	.25	.28	.36	.36	.36	.44	.43	.44
atoms			.072			.096			.117
K_2O %	.24	.20	.22	.18	.22	.20	.23	.26	.25
atoms			.037			.035			.044
P_2O_5 %	none	none	none	.11	.09	.10	.02	trace	.01
atoms			none			.012			.001
H_2O %	not determined			not determined			not determined		
atoms			5.758			2.280			2.304
Total %	93.43	93.55	93.50	97.72	97.30	97.51	97.56	97.40	97.49

Table 1. Chemical Analyses and Atomic Calculations

Sample	Actin. #175		Ave.	Actinolite (Phillips & Hess - 1936)	Biotite (Phillips & Hess - 1936)
SiO_2 %	58.66	58.03	58.34	53.27	38.72
atoms			7.829	7.619	2.843
TiO_2 %	.01	none	.01	.06	.47
atoms			.001	.006	.026
Al_2O_3 %	.03	none	.02	4.92	18.39
atoms			.003	.829	1.591
Fe_2O_3 %	.59	.53	.56	1.43	1.05
atoms			.057	.154	.058
FeO %	2.23	2.34	2.28	6.78	9.54
atoms			.256	.811	.585
MnO %	.12	.08	.10	.20	.16
atoms			.011	.024	.010
MgO %	24.73	25.06	24.90	19.47	18.44
atoms			4.977	4.148	2.017
CaO %	9.85	10.22	10.04	11.70	trace
atoms			1.443	1.792	trace
Na_2O %	.19	.19	.19	.75	.68
atoms			.049	.208	.097
K_2O %	.06	.09	.08	.59	9.74
atoms			.014	.108	.912
P_2O_5 %	trace	none	trace	.11	.11
atoms			trace	.013	.007
H_2O %	not determined			.44	.69
atoms			3.114	.687	1.322
(Calculations of water atoms assume difference between chemical total and 100% to be water.)					
Total %	96.47	96.54	96.52	99.72	97.99

Table 1. Chemical Analyses and Atomic Calculations

Sample	Calculated Atomic Formula
Biotite #152	$(K,Na,Ca)_{.97}(Mg,Fe'',Fe''',Ti,Mn,Al)_{2.72}(Si,Al,P)_{4.00}O_{9.68}(OH)_{2.32}$
Biotite #153	$(\_\_\_\_\_)_{.92}(\_\_\_\_\_)_{2.73}(\_\_\_\_\_)_{4.00}O_{9.51}(OH)_{2.49}$
Biotite #157	$(K,Na)_{.92}(Mg,Fe'',Fe''',Ti,Mn,Al)_{2.92}(Si,Al,P)_{4.00}O_{9.96}(OH)_{2.04}$
Biotite #158	$(\_\_\_)_{.94}(\_\_\_\_\_)_{2.85}(\_\_\_\_\_)_{4.00}O_{9.74}(OH)_{2.26}$
Biotite #160	$(\_\_\_)_{.90}(\_\_\_\_\_)_{2.86}(\_\_\_\_\_)_{4.00}O_{9.82}(OH)_{2.18}$
Biotite #167	$(\_\_\_)_{.86}(\_\_\_\_\_)_{2.96}(\_\_\_\_\_)_{4.00}O_{9.94}(OH)_{2.06}$
Biotite #172	$(\_\_\_)_{.89}(\_\_\_\_\_)_{2.88}(\_\_\_\_\_)_{4.00}O_{10.11}(OH)_{1.89}$
Biotite #176	$(\_\_\_)_{.89}(\_\_\_\_\_)_{2.98}(\_\_\_\_\_)_{4.00}O_{10.00}(OH)_{2.00}$
Biotite #206	$(\_\_\_,Ca)_{.92}(\_\_\_\_\_)_{2.85}(\_\_\_\_\_)_{4.00}O_{9.89}(OH)_{2.11}$
Biotite #208	$(\_\_\_)_{.95}(\_\_\_\_\_)_{2.86}(\_\_\_\_\_)_{4.00}O_{9.94}(OH)_{2.06}$
Biotite #209	$(\_\_\_)_{.88}(\_\_\_\_\_)_{2.94}(\_\_\_\_\_)_{4.00}O_{9.83}(OH)_{2.17}$
Biotite #210	$(\_\_\_)_{.95}(\_\_\_\_\_)_{2.92}(\_\_\_\_\_)_{4.00}O_{10.11}(OH)_{1.89}$
Biotite (Phillips & Hess - 1936)	$(\_\_\_)_{1.01}(\_\_\_\_\_)_{3.14}(\_\_\_\_\_)_{4.00}O_{10.68}(OH)_{1.32}$
Actinolite #152	$(Ca,Na,K)_{2.15}(Mg,Fe'',Fe''',Ti,Mn,Al)_{5.36}(Si,Al,P)_{8.00}O_{22.44}(OH)_{1.56}$
Actinolite #154	$(\_\_\_\_\_)_{2.23}(\_\_\_\_\_)_{5.22}(\_\_\_\_\_)_{8.00}O_{22.33}(OH)_{1.67}$
Actinolite #159	$(\_\_\_\_\_)_{2.05}(\_\_\_\_\_)_{4.98}(\_\_\_\_\_)_{8.00}O_{21.76}(OH)_{2.24}$
Actinolite #161	$(\_\_\_\_\_)_{2.01}(\_\_\_\_\_)_{4.49}(\_\_\_\_\_)_{7.38}O_{18.24}(OH)_{5.76}$
Actinolite #166	$(\_\_\_\_\_)_{1.93}(\_\_\_\_\_)_{5.03}(\_\_\_\_\_)_{8.00}O_{21.72}(OH)_{2.28}$
Actinolite #171	$(\_\_\_\_\_)_{1.91}(\_\_\_\_\_)_{5.09}(\_\_\_\_\_)_{8.00}O_{21.70}(OH)_{2.30}$
Actinolite #175	$(\_\_\_\_\_)_{1.51}(\_\_\_\_\_)_{5.30}(\_\_\_\_\_)_{7.83}O_{20.89}(OH)_{3.11}$
Actinolite (Phillips & Hess - 1936)	$(\_\_\_\_\_)_{2.11}(\_\_\_\_\_)_{5.60}(\_\_\_\_\_)_{8.00}O_{23.31}(OH)_{.69}$

Table 2. Analyses of Haplogranite, G-1, W-1, and Biotite #210

Sample	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	FeO	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	H_2O	Total
Haplo-granite	72.25	none	16.13	none	none	none	.84	1.79	3.37	5.76	none	none	100.14
	72.88	none	16.40	none	none	none	.77	1.73	3.30	5.76	none	none	100.84
Ave.	72.56	none	16.26	none	none	none	.81	1.76	3.34	5.76	none	none	100.49
Standard Composition (Fairbairn & Schairer - 1952)	72.64	none	15.78	none	none	none	.80	1.82	3.19	5.76	none	none	99.99
G-1	72.74	.24	14.40	1.06	.86	.02	.37	1.32	3.37	5.56	.09	.33	100.36
	72.88	.23	14.41	1.00	.90	.02	.33	1.30	3.53	5.67	.13	.35	100.75
Ave.	72.81	.24	14.40	1.03	.88	.02	.35	1.31	3.45	5.62	.11	.34	100.56
35 Lab. Average (Fairbairn - 1953)	72.36	.25	14.44	.93	.99	.03	.39	1.41	3.25	5.42	.09	.37	99.92
W-1	52.92	1.09	14.62	1.63	8.61	.16	6.74	10.74	2.32	.70	.18	.55	100.26
	53.42	1.09	14.80	1.56	8.68	.15	6.82	10.76	2.32	.72	.21	.51	101.04
Ave.	53.17	1.09	14.71	1.59	8.64	.16	6.78	10.75	2.32	.71	.19	.53	100.64
35 Lab. Average (Fairbairn - 1953)	52.34	1.10	15.07	1.50	8.71	.17	6.63	10.96	2.00	.63	.13	.56	99.79
Biotite #210	40.56	.05	18.65	1.29	3.40	.08	21.17	none	.28	10.24	.09	n.d.	95.81
(author)	39.90	.04	19.14	1.40	3.33	.09	21.75	none	.28	10.06	.08	n.d.	96.07
Ave.	40.23	.05	18.90	1.35	3.37	.09	21.46	none	.28	10.15	.09	n.d.	95.97
Biotite #210; Commer-cial Lab. A	31.46	.08	21.86	see below	.07	22.15	.05	.53	10.05				
	Also reported: Total iron to Fe_2O_3 - 5.83, Fluorine - 5.64, Loss on ignition (1000°C.) - 4.38												102.10
Biotite #210; Commer-cial Lab. B	37.94	.02	20.52	see below	.12	21.54	.35	1.30	10.48	trace			
	Also reported: Total iron to FeO - 2.78, Fluorine - .25, Loss on ignition - 4.63, Li_2O - .26, Boron - .005, Barium - .03, Beryllium - .002, and Molybdenum - .02.												100.25

Table 3. Published Analyses of Contact Biotite and Actinolite Samples

Sample	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	FeO	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	H_2O	Total
Biotite	39.08		15.38		7.12	.31	23.58		2.63	7.50		2.24	98.60
Also reported: Fluorine - .76													
(Emerson - 1895, Chester, Mass.; Contact of plagioclase vein in talc-chlorite seam)													
Biotite	39.55		15.95		7.80		22.25		10.35			4.10	100.00
(Emerson - 1895, Chester, Mass.; Contact of plagioclase vein in talc-chlorite seam)													
Biotite	40.80	.05	13.16	1.84	2.16		26.72	.46	2.44	7.95		2.95	101.42
Also reported: Fluorine - 2.89													
(v. Eckermann - 1925, Mansjo, Sweden; Pegmatite-limestone contact)													
Biotite	37.39	2.48	20.17	2.08	14.32		9.80	.51	.87	8.56		3.03	100.88
Also reported: Fluorine - 1.67; Cr_2O_3 - .02													
(Barth - 1936, Dutchess County, N.Y.; Pegmatite-schist contact)													
Biotite	38.72	.47	18.39	1.05	9.54	.16	18.44	trace	.68	9.74	.11	.69	97.99
Also reported: Fluorine - present but not determined													
(Phillips & Hess - 1936, Chester, Vt.; Serpentine-schist contact)													
Biotite	42.02	1.35	18.75	.66	8.29	.27	9.55	.93	.73	8.54		2.60	101.55
Also reported: Fluorine - 4.34; Li_2O - 1.20; Rb_2O - 1.85; Cs_2O - .47													
(Hess & Stevens - 1937, Kings Mt., N.C.; Mica schist-pegmatite contact)													
Biotite	39.97	2.64	17.51	2.26	14.81	.22	8.45	none	.45	8.48		2.80	104.01
Also reported: Fluorine - 3.17; Li_2O - .65; Rb_2O - 1.48; Cs_2O - 1.12													
(Hess & Stevens - 1937, Tin Mt., S.D.; Mica schist-pegmatite contact)													
Actinolite	53.27	.06	4.92	1.43	6.78	.20	19.47	11.70	.75	.59	.11	.44	99.72
(Phillips & Hess - 1936, Chester, Vt.; Serpentine-schist contact)													

to temperature and pressure, it can be partially tested with respect to composition. This has been the purpose of the chemical analyses. This section is included to indicate briefly the theoretical basis for the experimental work.

We will define our system of interest to be made up of a small length (a few feet, for example) of the two monomineralic bands. The system would be centered at the contact between the two bands and would include all material on either side of the contact which had been altered by interaction across the contact. The necessary conditions which must exist for equilibrium during the formation of the minerals are 1) that the temperature of all phases (minerals) be equal; 2) that the pressure on all phases be equal; and, 3) that the chemical potential of each component be the same in all phases in which it is present. By dealing with a small system the equal pressure requirement can be assumed to have existed. A temperature gradient perpendicular to the contact may have existed at the time of formation of the banding. If it did, the composition of the minerals might reflect this by exhibiting a compositional gradient perpendicular to the contact. We will assume that both the temperature and pressure requirements have been satisfied.

Let us consider the biotite grains in the biotite band. These are different examples of the same phase, namely: $K(Mg,Fe)AlSi_3O_{10}(OH)_2$. In particular we are interested in the Mg/Fe ratio of the different grains. If we assume that the magnesium and iron ions in a biotite grain represent an ideal mixture, i.e., that either

ion can substitute equally well in the structure, then the chemical potentials of magnesium and iron in the grain can be expressed as:

$$\mu_{Fe}^{\alpha} = \mu_{Fe}^{\circ} + RT \log \bar{n}_{Fe}^{\alpha}$$

and:

$$\mu_{Mg}^{\alpha} = \mu_{Mg}^{\circ} + RT \log \bar{n}_{Mg}^{\alpha}$$

where μ_{Fe}° refers to the chemical potential of iron in the pure state, μ_{Mg}° to the chemical potential of magnesium in the pure state, and $\bar{n}_{Fe}^{\alpha}$ refers to the mole fraction of iron based on the total number of moles found in six-fold co-ordination, $\bar{n}_{Mg}^{\alpha}$ to the mole fraction of magnesium on the same basis.

The assumption of ideal behavior is necessary to avoid use of activity coefficients, which give a quantitative measure of the discrepancies between actual behavior of a mixture and so-called "ideal" behavior. Ideal behavior is defined by certain thermodynamic relationships and represents the simplest possible behavior. We have very little information on the degree of ideality of mineral solid solutions, and no numerical values for the pertinent activity coefficients. The assumption of an ideal mixture for the magnesium and iron ions in biotites is thus an approximation. The two ionic radii do not differ greatly in size, a necessary condition for ideal behavior.

Assuming equilibrium has been attained in the biotite band, the necessary conditions of this state require that for two biotite grains: $\mu_{Fe}^{\alpha} = \mu_{Fe}^{\beta}$ and $\mu_{Mg}^{\alpha} = \mu_{Mg}^{\beta}$.

Therefore $\bar{n}_{Fe}^{\alpha} = \bar{n}_{Fe}^{\beta}$ and $\bar{n}_{Mg}^{\alpha} = \bar{n}_{Mg}^{\beta}$.

Thus, if we could analyze two separate biotite grains which have been formed in equilibrium with each other, the mole fraction of iron in each grain should be the same, as should be the mole fraction of magnesium. Naturally, in actual chemical analysis, a number of grains from one sample must be compared with a number of grains from another sample.

However other elements, such as aluminum, may substitute for iron and magnesium, and these other elements may not behave ideally with respect to each other. In this case, even though

$$\mu_{Fe}^{\alpha} = \mu_{Fe}^{\beta} \text{ and } \mu_{Mg}^{\alpha} = \mu_{Mg}^{\beta}, \quad \bar{n}_{Fe}^{\alpha} \neq \bar{n}_{Fe}^{\beta} \text{ and } \bar{n}_{Mg}^{\alpha} \neq \bar{n}_{Mg}^{\beta}.$$

For this situation the mole fractions of iron and magnesium should be based on the total number of moles of iron and magnesium only. A convenient method of comparison then is mole fraction ratios, with equilibrium requiring: $\bar{n}_{Mg}^{\alpha} \Big/ \bar{n}_{Fe}^{\alpha} = \bar{n}_{Mg}^{\beta} \Big/ \bar{n}_{Fe}^{\beta}$.

Thus attainment of equilibrium can still be checked by measuring the Mg/Fe ratios in biotite grains. In actual calculations a basis of 12(O,OH) atoms can be used to compute the number of Mg and Fe atoms represented by a given analysis, and the ratio of these numbers can be used as the equilibrium check.

Similar reasoning could be used to test attainment of equilibrium in a large homogeneous mass of rock, provided the constant temperature and constant pressure requirements can be assumed to have been fulfilled. Thus the Mg/Fe ratio would be a constant in biotite grains scattered through a pelitic schist which had formed

under equilibrium conditions. A systematic change in the ratio in a particular direction might indicate a temperature gradient or metasomatic movement of material. It is essential in studying metasomatism to check a rock for variations in mineral chemical composition rather than for variations in its bulk composition. Readers interested in a discussion of equilibrium in metasomatic processes are referred to a paper by Thompson (1959).

DISCUSSION OF RESULTS

Precision and Accuracy

A discussion of advantages and results obtainable in rapid analysis of silicate rocks is given by Shapiro and Brannock (1956). Mercey (1956) presents a detailed study of the accuracy and precision of the rapid methods as applied to rock analysis. He concludes that the accuracy and precision of these methods compares favorably with results obtainable by conventional methods. The author concurs with this conclusion, provided duplicate samples are analyzed and each sample is run in duplicate. The latter qualification adds relatively little to the total time required for the rapid techniques, and is a necessity since aberrant results do occur frequently enough to be significant. The concurrence applies only to analyses of samples with element concentration ranges of common igneous rocks. A further qualification must be made for the application of these methods to mineral analysis. In this case unusually high or low concentrations of various elements occur, and results comparable to those of conventional methods may not be obtainable.

The calculated atom values given here show good agreement between the chemical analyses and the requirements of the crystal chemistry of micas and amphiboles. For example, the sum of the cations of an amphibole should fall between fifteen and sixteen, and this value varies from 14.96 to 15.51 for five of the analyzed actinolites. The total for #175 is 14.64 and for #161, 13.88. The low summation of the chemical analysis of #161 indicates the probable presence of another major constituent. The analyses of the three standards provided another check on the mineral analyses, with results for the standards being similar to those obtained by conventional methods. The analyses of biotite #210 by the two commercial laboratories show wide disagreement in the results for several elements and neither laboratory shows close agreement with the author's values. This points out the importance of evaluating a mineral analysis by a crystal chemistry check.

General Results

Not unexpectedly, a rather large variation occurs in the composition of the two minerals. The major element variations in the biotites are SiO_2: 37.93 - 42.05%; Al_2O_3: 14.26 - 19.22%; FeO: 2.90 - 7.96%; Fe_2O_3: 1.13 - 3.04%; MgO: 17.98 - 24.42%; and K_2O: 8.96 - 10.31%. In the actinolites the variations are SiO_2: 51.33 - 58.34%; Al_2O_3: .02 - 7.70%; FeO: 2.28 - 3.90%; Fe_2O_3: .56 - 1.74%; MgO: 19.49 - 24.90%; and CaO: 10.04 - 13.34%. These variations, most of which are too great to be attributed to experimental error, indicate that only one or two analyzed specimens are an inadequate sample of the composition of such contact minerals.

Biotite samples #167 and #172 were taken where pelitic schist replaces quartz diorite as wall rock of the bands. The only difference in chemical composition of these samples is their high content of TiO_2 as compared to that of the other biotites. In the case of #172, which has 1.14% TiO_2, the color and index of the mineral differ from the other biotites, apparently because of the titania content. No change in titania content is shown by the co-existing actinolite samples of these two biotites.

High contents of CaO and P_2O_5 in biotite samples #152 and #153 would seem to indicate appreciable apatite impurity. However the samples were carefully checked by immersion in refractive index liquids and no apatite was found.

Biotite-Actinolite Pairs

A schematic representation of the relation of the analyzed biotite-actinolite pairs to the contact between the two monomineralic bands is shown in Figure 3. Figure 4 shows the variation in composition with respect to the contact between the two bands. As already pointed out, the four mineral pairs of Figure 4 do not actually come from one line across the two bands. Each pair is from a different point along the contact, with one pair from another contact, as has been shown in Figure 2. It is likely that Figure 4 represents the kind of variation which exists perpendicular to the contact. This is seen as culminations and depressions in the element content of the two minerals in the vicinity of the contact. A similar result was found by Dennen (1951) at the contact between certain intrusives and their wall rock. He reported:

Representation of Location of Analyzed Biotite-Actinolite Pairs With Respect to Contact Between The Two Monomineralic Bands

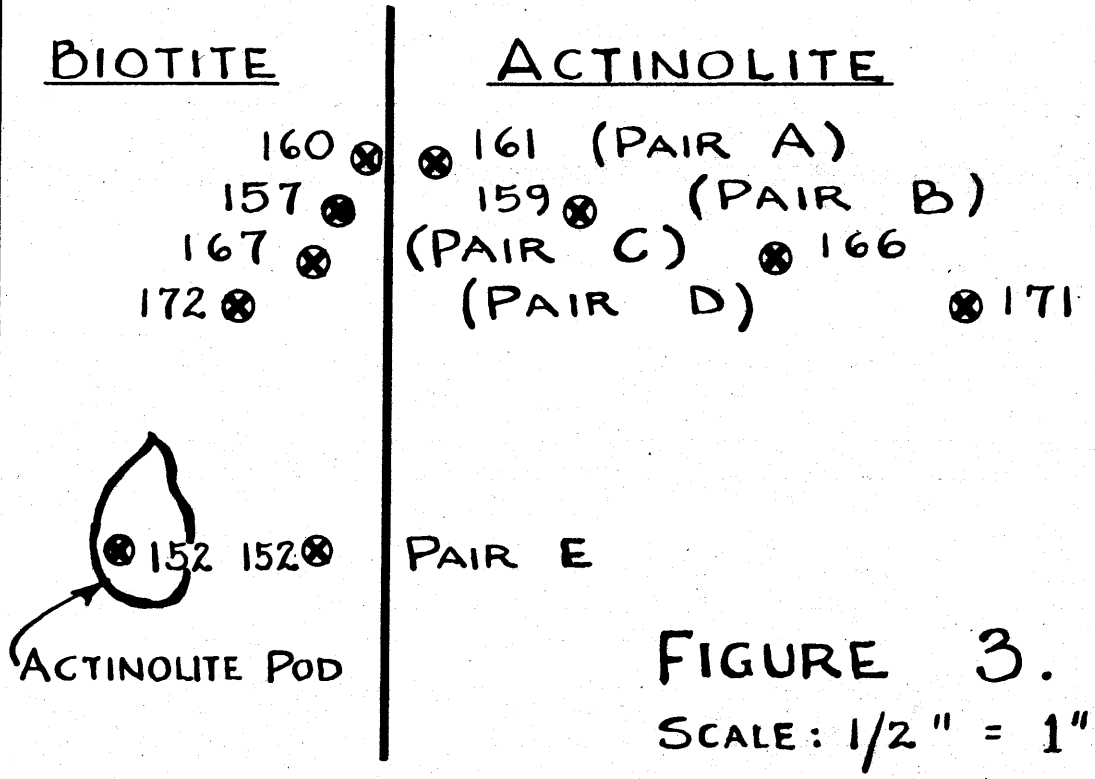
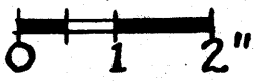


FIGURE 3.

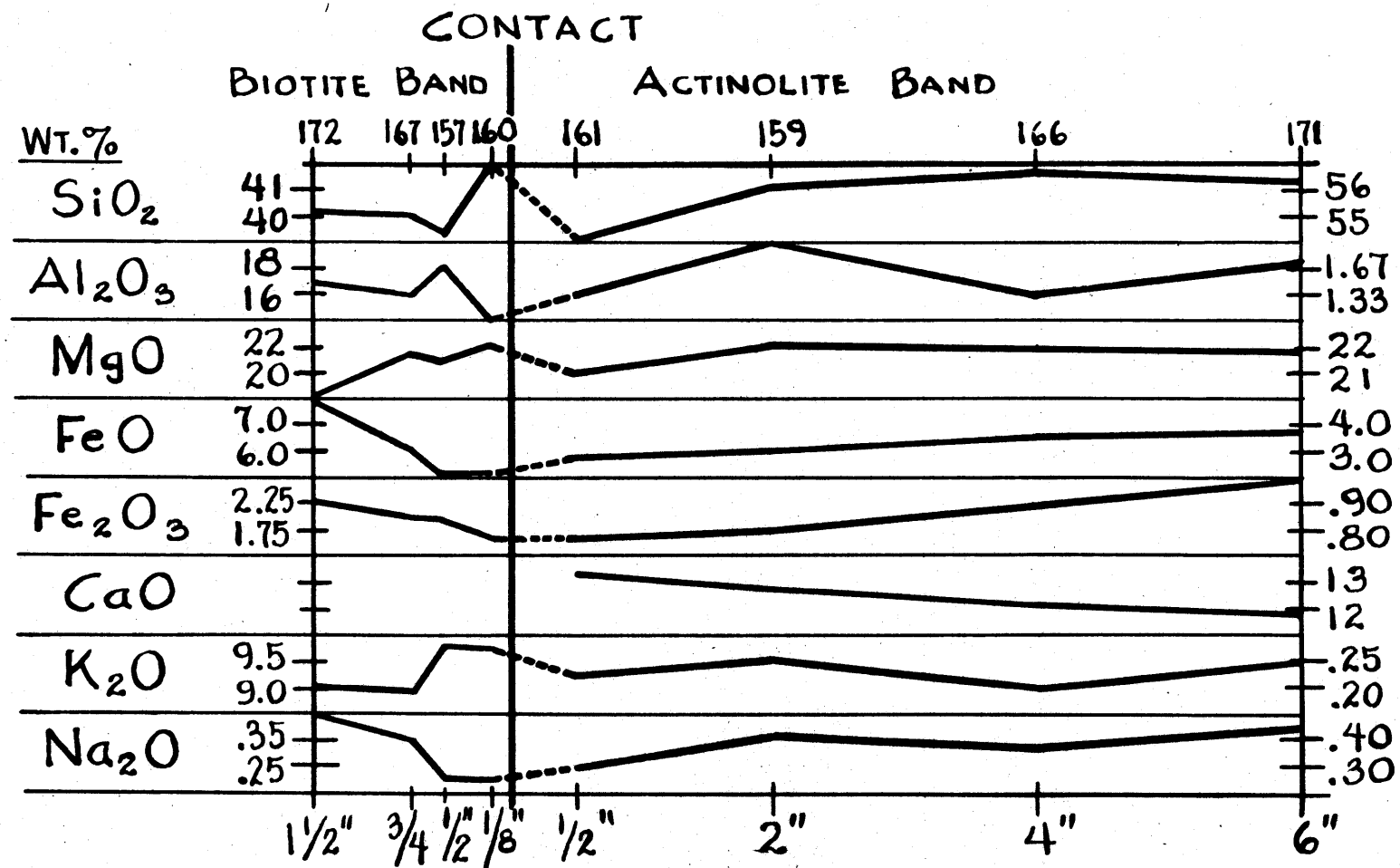
SCALE: 1/2" = 1"



PAIR A: (APPROXIMATE DISTANCE APART PERPENDICULAR TO CONTACT) 5/8"

PAIR B: 2 1/2" PAIR C: 4 3/4"

PAIR D: 7 1/2" PAIR E: 2"



MINERAL COMPOSITIONAL VARIATION WITH RESPECT TO BIOTITE – ACTINOLITE CONTACT

FIGURE 4.

"highly irregular variations are found when the intrusive and wall rock were compositionally different and steep thermal gradients existed. These latter variations may show geochemical culminations and depressions that are wave-like in form and die out in both directions from the contact zone." This work indicates that the variations found by Dennen in contact rocks exist also in contact minerals.

Figures 5 and 6 show the relationship of the Si-Al and Mg-Fe atomic ratios of the co-existing biotite-actinolite pairs. It is interesting to note that Pair E, which was taken at the contact of an actinolite pod in the biotite band, and therefore represents a definite disequilibrium situation, has a much different slope in both diagrams as compared to the other pairs, which were taken across the main contact between the two bands. Assuming that Mg and Fe have been introduced from the serpentine-marble, and Si and Al from the quartz diorite-schist, the change in slope of the pairs away from the main contact indicates silica to be more mobile than aluminum, and iron to be more mobile than magnesium. The variation in the Mg/Fe ratios in the two minerals indicates a degree of chemical disequilibrium in the bands, since it has been shown that attainment of equilibrium requires that the Mg/Fe ratios in different samples of the same mineral be similar. (See Fig. 7).

CONCLUSIONS

The calcareous rocks probably formed by metamorphism of dolomite rock containing varying amounts of silica and argillaceous impurity. Possible resulting rocks would include forsterite,

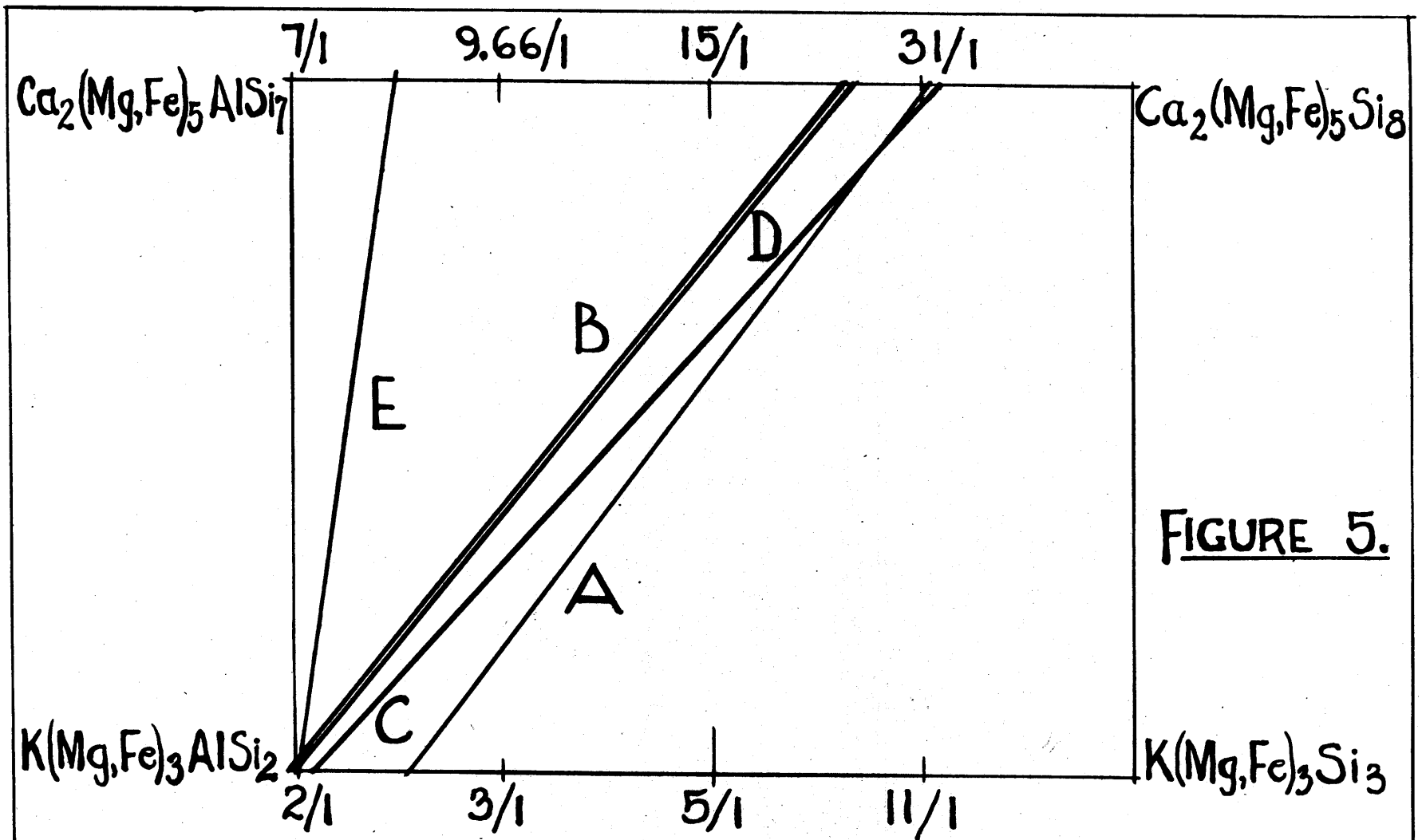


FIGURE 5.

Si-Al ATOMIC RATIOS FOR CO-EXISTING BIOTITE – ACTINOLITE SAMPLES

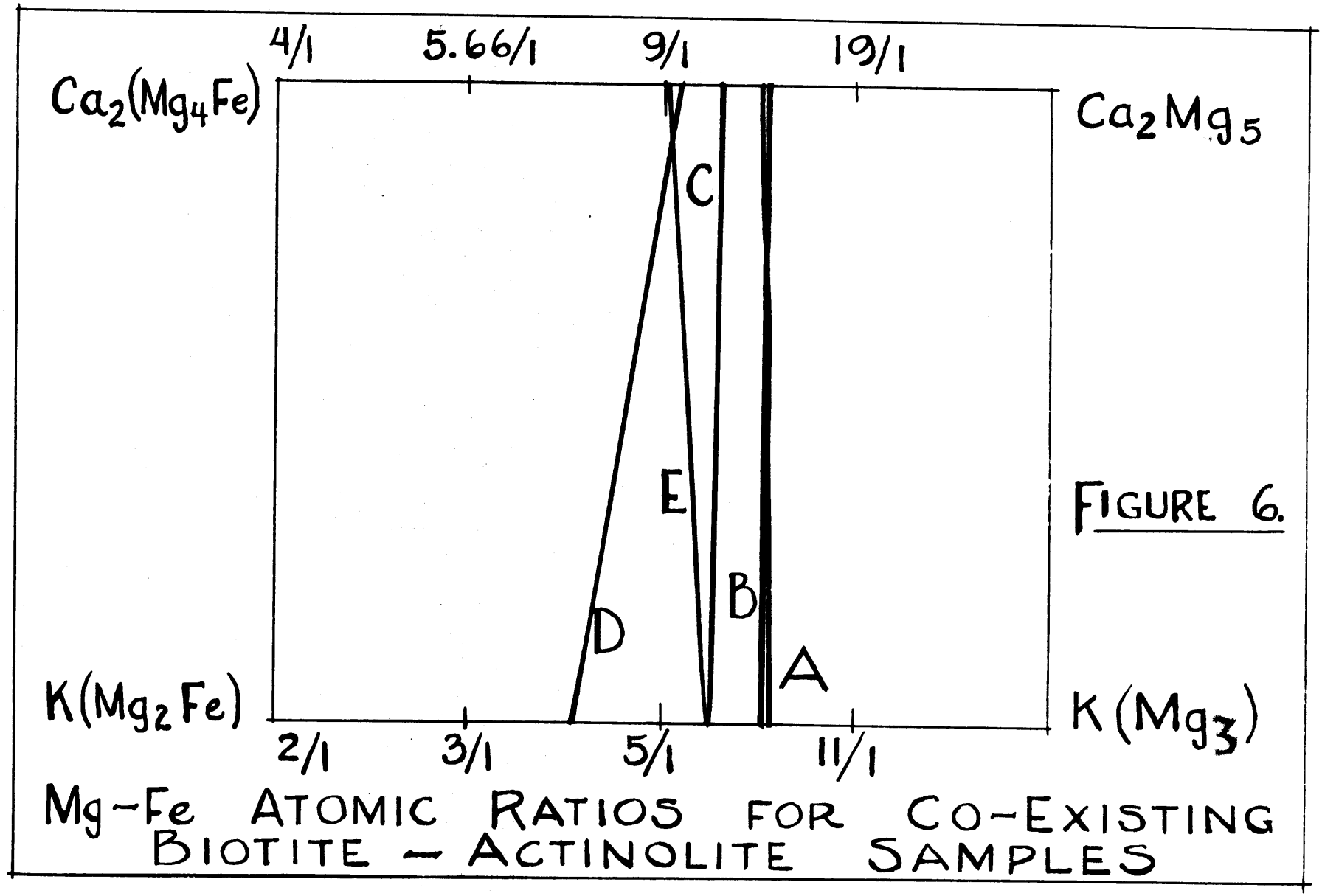


FIGURE 6.

Mg-Fe ATOMIC RATIOS FOR CO-EXISTING BIOTITE – ACTINOLITE SAMPLES

	PAIR A ※160	PAIR B ※157	PAIR C ※167	PAIR D ※172	PAIR E ※152	
Mg	2.338	2.206	2.273	1.917	2.121	BIOTITE
Mg/Fe	7.96	7.43	6.40	4.03	6.39	BIOTITE
Fe	.294	.297	.355	.475	.332	BIOTITE
BIOTITE Mg/Fe / ACTINOLITE Mg/Fe	.622	.567	.569	.393	.699	
Mg	4.075	4.506	4.495	4.482	4.180	ACTINOLITE
Mg/Fe	12.81	13.10	11.24	10.25	9.14	ACTINOLITE
Fe	.318	.344	.399	.437	.457	ACTINOLITE
	※161	※159	※166	※171	※152	

FIGURE 7.

CALCULATED Mg/Fe ATOMIC RATIOS IN BIOTITE-ACTINOLITE PAIRS

diopside, or actinolite marble, as well as various amphibolite and calc-silicate rocks. Addition of water from an intrusive or by squeezing of surrounding pelitic sediments would allow serpentine to form by replacement of fosterite, actinolite, and dolomite.

The monomineralic bands appear to be the result of very local reaction at the contact of the intrusive quartz diorite with the serpentine-marble. The chemical results show a large variation in the composition of the minerals. This fact, and the variation in the Mg/Fe ratios of the two minerals, indicates that thermodynamic equilibrium had not been reached at the time of formation of the bands. This is not an unexpected result, since minerals in such contact areas would be formed rapidly, without time for adjustments in distribution of elements, such as might occur in a slowly cooling magma or in regional metamorphism.

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Additional Note

Since completion of this manuscript the author has come across a recent publication by Kretz (1959). Kretz presents sixty-two spectrochemical analyses of co-existing biotite, garnet, and hornblende from Grenville gneisses of Quebec and discusses the results in terms of phase equilibrium. In testing the attainment of thermodynamic equilibrium he emphasizes use of the distribution coefficient for a given element between different minerals. It is concluded that a close approach to equilibrium was reached in these rocks.

Kretz, R. (1959), Chemical Study of Garnet, Biotite, and Hornblende from Gneisses of Southwestern Quebec with Emphasis on Distribution of Elements in Coexisting Minerals, Jour. Geol., v. 67, p. 371-402.

References

Brownlow, A. H. (1960), Serpentine and Associated Rocks and Contact Minerals near Westfield, Massachusetts: Ph.D. Thesis, Massachusetts Institute of Technology.

Chidester, A. H. et al. (1951), Talc Investigations in Vermont: U. S. Geol. Surv., Circular 95.

Dennen, W. H. (1951), Variations in Chemical Composition Across Igneous Contacts: Geol. Soc. America Bull., v. 62, p. 547-558.

DuRietz, T. (1935), Peridotites, Serpentines, and Soapstones of Northern Sweden: Geo. Fören. Förh., v. 57, p. 133-260.

Emerson, B. K. (1898), Geology of Old Hampshire County, Massachusetts: U. S. Geol. Surv., Mon. 29.

Fairbairn, H. W. (1953), Precision and Accuracy of Chemical Analysis of Silicate Rocks: Geochim. Cosmochim. Acta, v. 4, p. 143-156.

_____, and Schairer, J. F. (1952), A Test of the Accuracy of Chemical Analysis of Silicate Rocks: Am. Mineral., v. 37, p. 744-757.

Gillson, J. L. (1927), Origin of the Vermont Talc Deposits: Econ. Geol., v. 22, p. 246-287.

Haapla, P. (1936), On the Serpentine Rocks in Northern Karelia: Comm. Geol. Finlande, Bull., v. 114.

Hall, A. J. (1941), The Relation Between Color and Chemical Composition in the Biotites: Am. Mineral., v. 26, p. 29-41.

Heinrich, E. W. (1946), Studies in the Mica Group; The Biotite-Phlogopite Series: Am. Jour. Sci., v. 244, p. 836-848.

_____, et al. (1953), Studies in the Natural History of Micas: Univ. of Michigan Engineering Research Inst.

MacDonald, G. A. (1941), Progressive Metasomatism of Serpentine in the Sierra Nevada of California: Am. Mineral., v. 26, p. 276-287.

Mercey, E. L. P. (1956), The Accuracy and Precision of "Rapid Methods" of Silicate Analysis: Geochim. Cosmochim. Acta, v. 9, p. 161-173.

Nockolds, S. R. (1947), The Relation Between Chemical Composition and Paragenesis in the Biotite Micas of Igneous Rocks: Am. Jour. Sci., v. 245, p. 401-420.

Pabst, A. (1942), The Mineralogy of Metamorphosed Serpentine at Humphreys, Fresno County, California: Am. Mineral., v. 27, p. 570-585.

Phillips, A. H., and Hess, H. H. (1936), Metamorphic Differentiation at Contacts Between Serpentine and Siliceous Country Rocks: Am. Mineral., v. 21, p. 333-362.

Read, H. H. (1934), On Zoned Associations of Antigorite, Talc, Actinolite, Chlorite, and Biotite in Unst, Shetland Islands: Mineral Mag., v. 23, p. 519-540.

Riley, J. P. (1958), The Rapid Analysis of Silicate Rocks and Minerals: Anal. Chim. Acta, v. 19, p. 413-428.

Shapiro, L., and Brannock, W. W. (1956), Rapid Analysis of Silicate Rocks; A Contribution to Geochemistry: U. S. Geol. Surv., Bull. 1036-C, 56 p.

Thompson, J. B., Jr. (1959), Local Equilibrium in Metasomatic Processes: Researches in Geochemistry, P. H. Abelson, Editor, John Wiley & Sons, Inc., N. Y., N. Y., 511 p.

PART II

Chapter I. The Serpentine Problem

HISTORICAL REVIEW

The serpentine rocks to be discussed in the following pages are not those found in layered intrusions associated with much greater masses of more siliceous rock, and formed by alteration of peridotite and dunite segregations. To be considered here are those bodies which occur as lenticular masses, generally conforming to the schistosity of the enclosing rocks, in belts following mountain chains and island arcs, and in association with rocks of geosynclinal origin, but with no significant related intrusives. They are often referred to as peridotite, but are usually serpentinized at least in part.

Early geologic studies on serpentine bodies and their manner of formation had led, by the beginning of the twentieth century, to a general conclusion of derivation from ultramafic intrusives. These studies linked serpentine rocks to anhydrous magnesium rocks and gave outlines of the process of serpentinization, discussing the minerals affected and the types of serpentine formed. The conclusion thus developed has continued to be accepted to the present time, with no importance attached to rare cases where small amounts of serpentine have been formed by hydration of metamorphic silicates in altered magnesian limestone sediments. However no overall agreement on the source of the water of serpentinization has emerged. This problem and the tectonic and historical significance of regional serpentine belts have been the main subjects of more recent investigations.

Although local rock associations of serpentine bodies had been discussed prior to 1905, it was in that year that Steinmann (1905) pointed out the world-wide association of serpentine with spilites and cherts in orogenic areas. In the following years a great amount of study was devoted to this group of rocks, in particular with respect to their relationship to a general igneous cycle in orogenic development. The serpentines and altered volcanic rocks were discussed in the literature as ophiolites by continental petrologists, while in English literature they were referred to as the green rocks. These studies led to the assignment of the basic volcanic rocks and serpentines to the early stages of the development of mountain systems.

A complete review of the literature on the origin of serpentine has been given by Benson (1918). He listed four possible sources for the water of serpentinization: 1) the invaded rock; 2) a mixture of magmatic and meteoric ground water; 3) the ultramafic magma itself; and 4) other intrusions. From the field data he concluded that serpentinization is an autometasomatic process, with the water supplied by the magma itself, and alteration occurring late in the history of the intrusion. Benson (1926) extended his review of the literature to include both basic and ultramafic rocks, and clearly related the type of igneous rock produced and the tectonic conditions existing in a given area. He listed seven major modes of occurrence of intrusive mafic and ultramafic rocks, and refers to ultramafics associated with orogenic zones as the "alpine type," a term widely used by later writers.

In the past thirty years most geologists have appealed to one of two alternative means of serpentinization, the autometasomatic hypothesis of Benson, or the solid state intrusion mechanism outlined by Bowen (1928). Bowen pictured ultramafic intrusions as consisting of a mass of olivine crystals with only a small amount of interstitial magmatic or water vapor. This mass would represent a crystal fraction formed by differentiation from basaltic magma. The initial movement of the intrusion would be due to external forces, producing a certain amount of deformation as the flow continued. The invading mass would then absorb water from wet sediments, resulting in increased mobility and serpentinization. Various rock types would develop depending on the amount of water absorbed and on whether some pyroxene was included in the original "crystal mush."

Hess (1933) discusses and separates two distinct types of alteration of ultramafics, serpentinization and steatitization (talc formation). He shows that these may occur separately or together, with steatitization always younger than serpentinization and unrelated to it in any way. Serpentinization is believed to be caused by late-magmatic autometamorphism and steatitization to hydrothermal solutions, often traced to acid intrusions.

Hess (1938) also has summed up the field evidence for the existence of a completely liquid ultrabasic magma. This he postulated to be a highly aqueous liquid approximating serpentine in composition. With cooling temperature olivine would separate out, followed at lower temperature by reaction of the olivine with a

large volume of residual aqueous silicic acid solution to form serpentine. This suggestion was made to solve the problem raised by laboratory studies by Bowen and Schairer (1936), which indicated that a peridotite "magma" could exist only above about 1600°C, whereas field evidence on the contact effects of serpentinites indicated temperatures in the vicinity of 500°C. The "serpentine magma" hypothesis also solves the "equal-volume" problem. Field evidence indicates essentially no increase in volume due to serpentinization of ultramafics. A constant volume reaction between olivine and water requires extensive magnesia metasomatism of adjoining rocks, which is not found, or that great volumes of water be available for the alteration.

A further contribution to serpentine studies was made by Hess (1937, 1939) in pointing out the coincidence between serpentine belts and belts of negative gravity anomalies. He suggested that this may arise from thickening of the granitic crust in these zones, causing differential fusion of a lower, assumed peridotite layer, thus producing ultramafic magmas.

Another relationship has been most clearly detailed by Taliaferro (1943). He presented the field data and petrology of the glaucophane schists and associated ultramafics of California, which is representative of the many areas where these rocks occur together. The glaucophane schists are dense, soda-rich rocks which appear to have formed by introduction of soda and other elements into geosynclinal sediments and volcanics, often near ultramafic contacts. However, there are many serpentinite areas

which have no glaucophane rocks and vice versa, and the direct connection, if any, between the two types is still unknown.

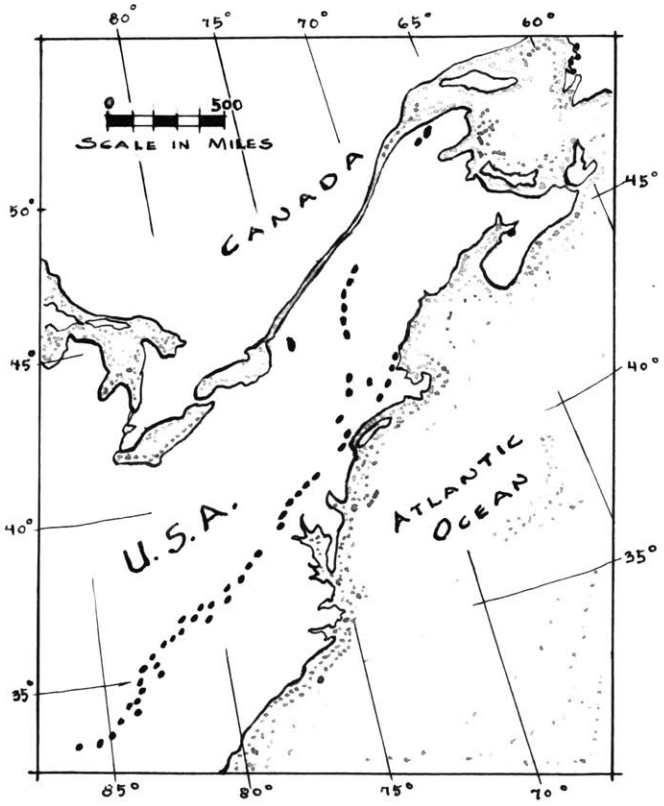
A significant contribution to the serpentine problem was made by Bowen and Tuttle (1949) in a laboratory study of the system MgO-SiO_2- H_2O. This system was studied at pressures exceeding 2000 atmospheres and at temperatures up to 1000°C, with no liquid phase found. They concluded that there is no possibility of the existence of a serpentine magma below 1000°C. Other results indicated that pure magnesian serpentine cannot exist above approximately 500°C; forsterite is stable in contact with water down to about 400°C, being then replaced by serpentine and brucite; iron-bearing olivine is stable to even lower temperatures. Bowen and Tuttle clearly point out that these results require that ultramafic bodies be intruded in the solid state, with serpentinization developed by introduction of solutions into a crystalline mass composed of anhydrous minerals. The "crystal mush" hypothesis of Bowen is amplified in light of the experimental results. It should be pointed out that the serpentine mineral dealt with by Bowen and Tuttle was chrysolite, whereas antigorite is more common in field occurrences.

Although the laboratory work of Bowen and Tuttle rules out the existence of a serpentine magma, Hess and other field geologists still stand by intrusion of ultramafics in a fluid state as best satisfying field evidence. They suggest that some factor is missing in the laboratory investigations. In a recent paper Hess (1955) restates the field objections to the Bowen-Tuttle

hypothesis. These include the occurrence of many serpentinites in areas where deformation has not been intense, lack of evidence of flow in the internal fabric of the majority of serpentinites, and no evidence of extensive water vapor, which Hess feels would be required for flow of the solid intrusion. He defines the present state of the serpentine problem as follows: "The facts can be summarized, and some conclusions drawn concerning the serpentinites of alpine mountain systems. But satisfying general conclusions which would account in one all-embracing theory for their origin, mode of emplacement, and subsequent serpentinization are not forthcoming at this time." (Hess, 1955, p. 392)

THE SERPENTINE BELT OF WESTERN MASSACHUSETTS

One of the longest and most continuous serpentine belts in the world runs parallel to the Appalachian axis from Newfoundland to Alabama in Eastern North America (Figure 1). This belt was first outlined by Pratt and Lewis (1905), although parts of it had been discussed in earlier literature. Hunt (1854) reviewed the regional geology of northeastern North America as known at that time, and noted the association of a north-south serpentine belt with limestone and dolomite. Earlier, Hitchcock (1841), in the first comprehensive study of the geology of Massachusetts, described many of the serpentine occurrences of the Massachusetts section of this belt, which crosses the western part of the state (Figure 2). The work of Hitchcock was extended and brought up to date by Emerson (1895, 1898, 1917).



THE APPALACHIAN SERPENTINE BELT
AFTER *PRATT AND LEWIS (1905)*

FIGURE 1.

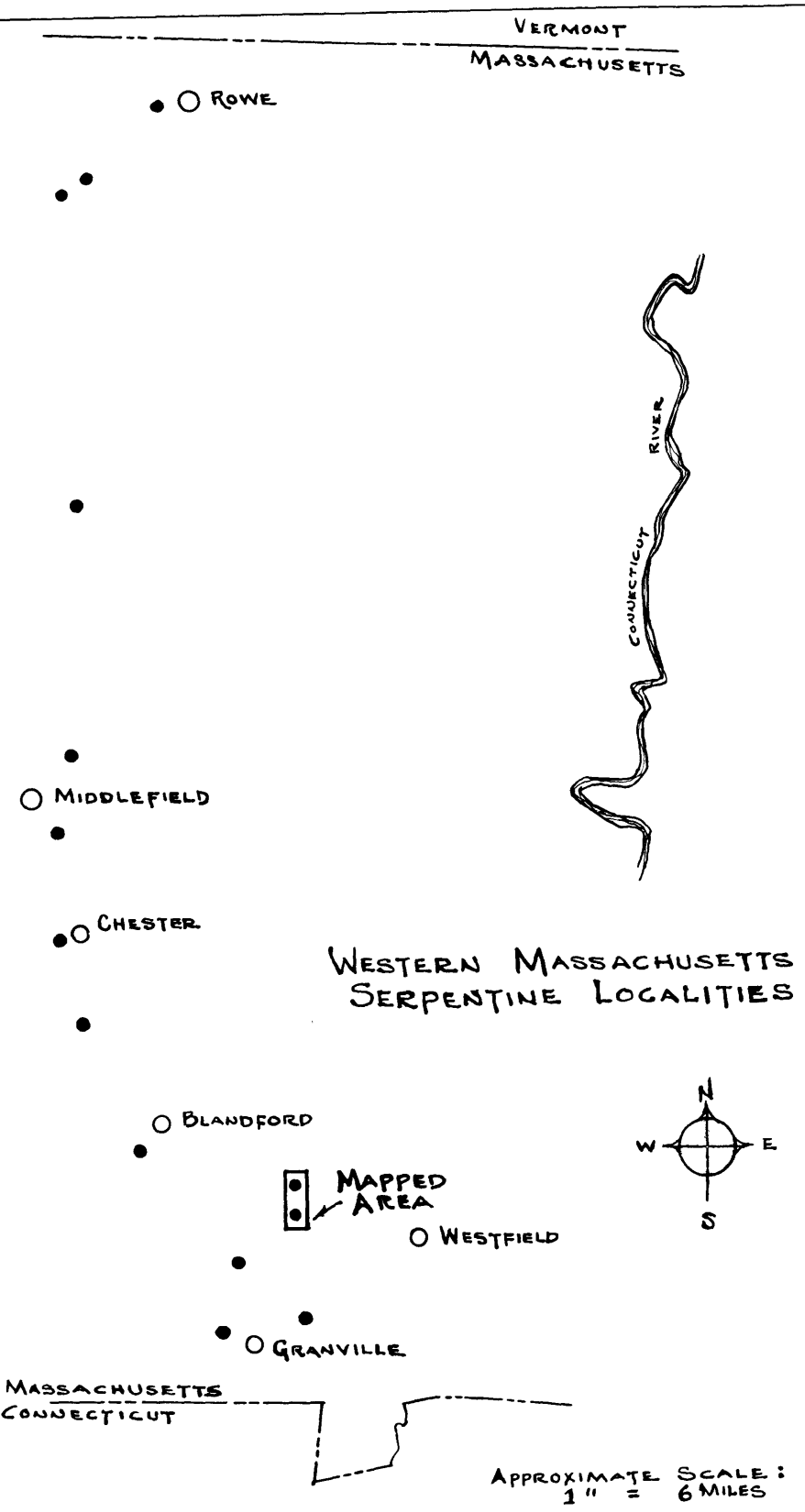


FIGURE 2.

AFTER EMERSON - 1898

The area on which the present research is based occurs in the Massachusetts section of the eastern North America serpentine belt (see Figure 2). The following summary of the more important features of this section, taken mostly from the work of Hitchcock and Emerson, is given to provide a regional picture of the western Massachusetts serpentines, and to show the many similarities of the separate occurrences. In particular, serpentine relationship to carbonate rocks and monomineralic contact features will be outlined. Serpentine is found in several places in Massachusetts east of the Connecticut River, but, with one exception, these will not be discussed here.

The physiography of western Massachusetts is characterized by old, strongly eroded mountain groups and ranges. These extend above a plateau-like upland, which is cut by narrow valleys and which slopes toward the coast of New England. The upland is part of an extensive erosion surface formed during the general peneplanation of all the Appalachian region at the beginning of the Cenozoic era. The topography has been slightly modified by glaciation and covered by a thin mantle of glacial drift. At least fourteen separate occurrences of one or more small serpentine bodies, associated with a thin amphibolite bed, run in a warped north-south band from Rowe and Leyden in northern Massachusetts to Granville in southern Massachusetts. These serpentine bodies vary in width from a few feet to several hundred feet and in length from tens of feet to a few miles, and occur in strongly metamorphosed early Paleozoic sediments, with the general strike of the serpentine paralleling the strike of the major rock groups.

The steeply dipping sediments are represented by mica schists and gneisses, with lesser limestone and dolomite marble. There are numerous granitic intrusions. Scattered through the altered sediments are thin beds of intercalated amphibolite.

Emerson (1898) noted the constant association of the serpentine bodies with a thin, discontinuous, epidote-hornblende-plagioclase amphibolite band, the Chester amphibolite. He found it to be 500 feet wide in the northern part of the state, widening to 3200 feet at Chester, and then thinning to the south. His mapping indicated the amphibolite and enclosing schists to be one conformable series of beds, with the amphibolite often occurring as several thin bands in the schists. The serpentine bodies were found to occur along the eastern border of the amphibolite, as were lenses of emery which occur at Chester. South of Chester, dolomitic marble becomes associated with the amphibolite and serpentine, and further south the proportion of marble increases and that of the amphibolite decreases.

The Chester area has received more study than the other serpentine localities because of the emery (corundum-magnetite) found there. Small scale mining of the emery occurred at various times between approximately 1860 and 1920. Generally the emery occurs as thin conformable veins in thin lenses of emery schist (chlorite, margarite, diaspore, epidote, tourmaline, magnetite, corundum) in the amphibolite, usually near its contact with serpentine. The amphibolite is composed of plagioclase, amphibole, epidote, and muscovite. At various places between the amphibolite

and the regionally enclosing mica schists are thin beds of talc and chlorite, or talc and serpentine, often with abundant carbonate. The amphibolite and other rocks seem to form a single major bed, usually separated from the mica schists by a few feet of chlorite. A few thin beds of amphibolite are also found nearby in the mica schists. All of the rocks of the area trend north-south. The total length of the emery occurrences is about seven miles.

Thin bands, one inch to over a foot wide, of combinations of biotite, actinolite, and chlorite separate the emery schist from the amphibolite and often fill cracks and fissures. The minerals in these openings may be associated with one or more of the following: margarite, diaspore, pyrite, and rutile. Tourmaline is in places abundant in the emery schist near the banding. At a few localities this banding separates emery schist from thin veins of fine-grained oligoclase, which sometimes have small crystals of corundum scattered through them. These feldspar veins with adjacent banding are also found in the serpentine-talc and talc-chlorite bodies. In some of these occurrences the contact minerals grow at right angles to the plane of contact with the feldspar or amphibolite, and are often very coarse-grained. Also found locally in the emery schist are small quartz segregations. Most of the above descriptive material is the result of a tour of the Chester area given the writer by David Wones, who has recently mapped parts of it. Mr. Wones has kindly allowed use of this material here.

The only other area in Massachusetts with similar geology is near Pelham, on the eastern side of the Connecticut River. (Emerson - 1898, p. 47) Here a granitic gneiss grades into an anorthite mass which is in contact with olivine-bronzite rock. At the contact is a biotite band from four inches to four feet thick. This band has nodules of hornblende, actinolite, and corundum wrapped in a chloritic material. The contact area also has allanite, rutile, apatite, and tourmaline. Within a few hundred feet of this contact are two large serpentine bodies.

The occurrence of contact mica bands at two serpentine quarries near Blandford is reported by Emerson (1898). At one of these a talc bed is separated by one inch of black mica and inch of hornblende-magnetite rock from a dike of granite. Bands of biotite and actinolite are found at the Westfield quarries and will be discussed in more detail in the next chapter. It is likely that this contact banding is a common feature of many of the Massachusetts serpentine localities, but it has been exposed only in those areas where quarrying or mining has been carried out.

The association of the serpentines of western Massachusetts with limestone and/or dolomite marble is of great interest, and appears to be a general feature. In many cases the carbonate is an accessory mineral or, where abundant, does not obviously represent an original sedimentary bed. Other occurrences, particularly in the southern part of the state, definitely appear to represent original sediments. Usually they appear as small beds in the vicinity of the serpentine, but sometimes an intimate mixture of serpentine and carbonate is found. Hitchcock (1841, p. 567)

reported three occurrences along the west branch of the Westfield River in the vicinity of Becket and Middlefield, where "it seems as if a part of the limestone itself were changed into serpentine." Similar relationships are found at Granville and at Westfield, where the serpentine occurs both as scattered lenses in a carbonate groundmass and as a massive gradation from pure carbonate. This type of rock, when polished, is known as verde antique marble. Continental petrologists often refer to it as ophicalcite. Hitchcock (1841, p. 569) also mentions an "augitic limestone," composed of dolomite and disseminated green augite, occurring at New Marlborough, Tyringham, and Sheffield, Massachusetts, and at Canaan, Connecticut. All of these localities are to the west of the main serpentine belt. No other verde antique localities are reported in western Massachusetts, but several occur further south in Connecticut.

Emerson reported that an irregular thickness of dolomitic limestone appears in the Chester amphibolite at Blandford, and gradually replaces the amphibolite to the south, while the limestone becomes pierced with large enstatite prisms and grades into a coarse "enstatite rock," which in turn grades into a verde antique marble or a black bastite serpentine. A point stressed by Emerson in 1898 was the lack of olivine in most of the serpentine bodies. He found abundant olivine at one of the quarries in Blandford, and some olivine pseudomorphs in Middlefield. The other bodies had no sign of olivine. This fact, combined with the dolomite-serpentine-amphibolite relationship found in the

southern part of the state, and with evidence of serpentine always occurring along the upper surface of the amphibolite, led Emerson to conclude in 1898 that all these rocks were originally a more or less impure ferruginous dolomitic limestone. The enstatite-serpentine rock he attributed to reaction of the limestone with heated siliceous solutions from the numerous granitic intrusions found in the southern part of the state, while the Chester emery deposits represented metamorphism of sedimentary limonite-gibbsite beds. He listed two features which might indicate an eruptive origin for the serpentine and amphibolite: 1) a relatively constant content of chromium (approximately .30% Cr_2O_3) and nickel (approximately .35% NiO) found in analyzed specimens from the serpentines and associated rocks; and 2) a close agreement in composition and structure of the amphibolite with rocks which have been proved to be derived from mafic eruptives. Chromite usually occurs as disseminated grains in the serpentine and, in some localities, as thin masses parallel to the stratification.

By 1917 Emerson had changed his mind with respect to the serpentine and now considered these bodies to be of igneous origin, although he still thought the Chester amphibolite to be of sedimentary origin. In particular he emphasized the constant nickel and chromium content of the serpentine. The emery at Chester he suggested to be a metamorphic reaction rim between igneous olivine and the amphibolite. The main factor which led him to this new view appears to be the work of Palache (1907). Palache (p. 491) found abundant olivine in thin sections of massive serpentine

from Chester and reported "in every detail the specimen is a typical peridotite...". He also found olivine occurring as a narrow vein in the massive serpentine. Emerson also mentions Martin (1898), who found olivine in thin sections and confirmed its presence by chemical analysis in a study of a small ultrabasic body occurring west of the serpentine belt near Cheshire.

With the exception of some general quadrangle mapping, no published work on the serpentines of western Massachusetts has appeared since 1917. The common features of these serpentines, as described in the references mentioned in the preceding pages, can be summarized as follows: 1) they generally occur as a single lens or group of lenses parallel to the structure of the surrounding rock, and are of small size, with both lengths and widths measured in terms of a few feet to a few miles; 2) they consist almost entirely of serpentine, with some talc and carbonate, and contain olivine in only a few localities; 3) they are almost always associated with thin beds of amphibolite, and often also with similar beds of marble, at some localities forming verde antique marble; 4) analyses of the serpentine form several localities indicate a small but constant content of chromium and nickel; and 5) at Blandford and Westfield, where serpentine contacts have been exposed, thin monomineralic banding has been found. Similar banding is found at the contacts of emery schist associated with serpentine at Chester.

CHAPTER II. FIELD GEOLOGY

HISTORICAL REVIEW

The earliest mention in scientific literature of serpentine in the vicinity of Westfield is a brief reference to it in a paper on a Southampton lead mine by Eaton (1819), which was published in the first volume of Silliman's Journal, later to become the American Journal of Science. Two years later Atwater (1821) gave a few details on a "hill of serpentine" at Westfield. Two main serpentine bodies about 3/4 mile apart are found near Westfield, and this paper probably referred to the northernmost occurrence. The first reference to the southern locality was made by Davis (1826) in a paper on the rocks and minerals of Westfield. A very brief description of serpentine and associated minerals at Westfield is given by Hitchcock (1841, pp. 617-18). No further notice of Westfield serpentine is found until the series of publications by Emerson (1895, 1898, 1917), mentioned earlier, and a short geological report by Crosby (1896).

Quarries had been opened at both [redacted] of the Westfield serpentine bodies by the time that Emerson and Crosby visited them. These quarries are now filled with water. To supplement the present field work, the relationships reported by Emerson and Crosby will be summarized here in some detail. Crosby's report, which can be found in the Westfield public library, dealt with the southern serpentine mass, where a quarry had been opened a few years earlier by the Westfield Marble and Sandstone Company. In the quarry, across a thickness of about one hundred feet, he found

three parallel varieties of rock from west to east: 1) massive black and green serpentine; 2) laminated grey and green verde antique dolomite marble; and 3) massive black and grey spangled dolomite marble consisting of a scattered network of pseudomorphous serpentine prisms in a groundmass of dolomite. An analysis of the marble indicated it to be, apart from the serpentine, a nearly pure dolomite. Crosby suggested formation of these rocks by intrusion of a basic dike next to an impure dolomite limestone bed followed by serpentinous alteration of both rocks.

Emerson's 1895 publication merely lists the various minerals occurring at Westfield. In the 1898 monograph (pp. 78-155), Emerson gives a detailed description of the two Westfield serpentine bodies, particularly the southern locality, on which he based his main conclusions on the "enstatite rock." This "enstatite rock" he found to be a regional feature, associated with serpentine, marble, and amphibolite, from Blandford to the southern Massachusetts border. He concluded that these rocks were all derived from an impure dolomite limestone, and extended this conclusion to the serpentine and amphibolite running north from Blandford to the Vermont border. In 1917 Emerson gave no further descriptive material for the Westfield occurrences, but, as discussed earlier, had changed his opinion on the origin of the serpentines of western Massachusetts, which he now felt were of igneous origin.

In 1898 Emerson described the northern Westfield serpentine body ("Atwater's ledge"), which had been operated as a quarry for some years by that time, as having distinct foliation, bordered

on the west by a biotite-hornblende schist, and composed of black serpentine with abundant green foliated bastite. In thin sections of this rock he found the serpentine structure to indicate formation from enstatite, and concluded that "the rock from which the serpentine was derived was a nearly or quite pure enstatite rock." An analysis of this serpentine is among those of western Massachusetts serpentines given by Emerson, who reported a nickel oxide content of .40% and a chromium oxide content of .33%.

At the southern quarry Emerson reported, as had Crosby, three distinct beds: 1) "black marble, like that of the old Atwater quarry,...a black enstatite rock...now in various stages of serpentine change;" 2) "black spotted marble...containing much shining tremolite and spotted with elongate crystals of the same black altered enstatite;" and 3) "the third bed, which connects the other two, is a thin...foliated...white marble with...serpentine." Emerson also noted a partial change of the central limestone bed into tremolite, and found actinolite bands cutting across both the enstatite and limestone. Besides evidence of serpentine replacement of enstatite, Emerson also found in thin section "every stage of the change of dolomite into a colorless, almost perfectly amorphous serpentine...and in many cases these serpentine grains retain perfectly the cleavage and repeated twinning planes of the dolomite."

Although the postulated enstatite had been completely changed to serpentine, Emerson found that "every stage of the change can be seen, from that in which the black enstatite is crushed into

black bands between layers of limestone, only a few bronzy cleavage surfaces remaining, to that in which the long, narrow bands and spots of the rich green serpentine, with their lighter centers, are the last remnants of the black serpentinized enstatites with their grey centers." Also, "transverse sections of crystals, from both the massive and implanted forms, show the prismatic and two pinacoidal cleavages of enstatite, marked with great regularity by bands of magnetite dust, even when the whole mass is completely changed to serpentine." Emerson found definite indications that the central band of limestone had been sheared, but examination of the black mottled marble, which showed no evidence of internal movements, led him to conclude that the serpentine pseudomorphs "must, then, have been formed since the erection of the beds into their present vertical position and during their penetration by the abundant granite dikes which cut them." The formation of the enstatite and serpentine he attributed to reaction between a sedimentary ferruginous dolomite and permeating siliceous solutions from regional intrusive granites.

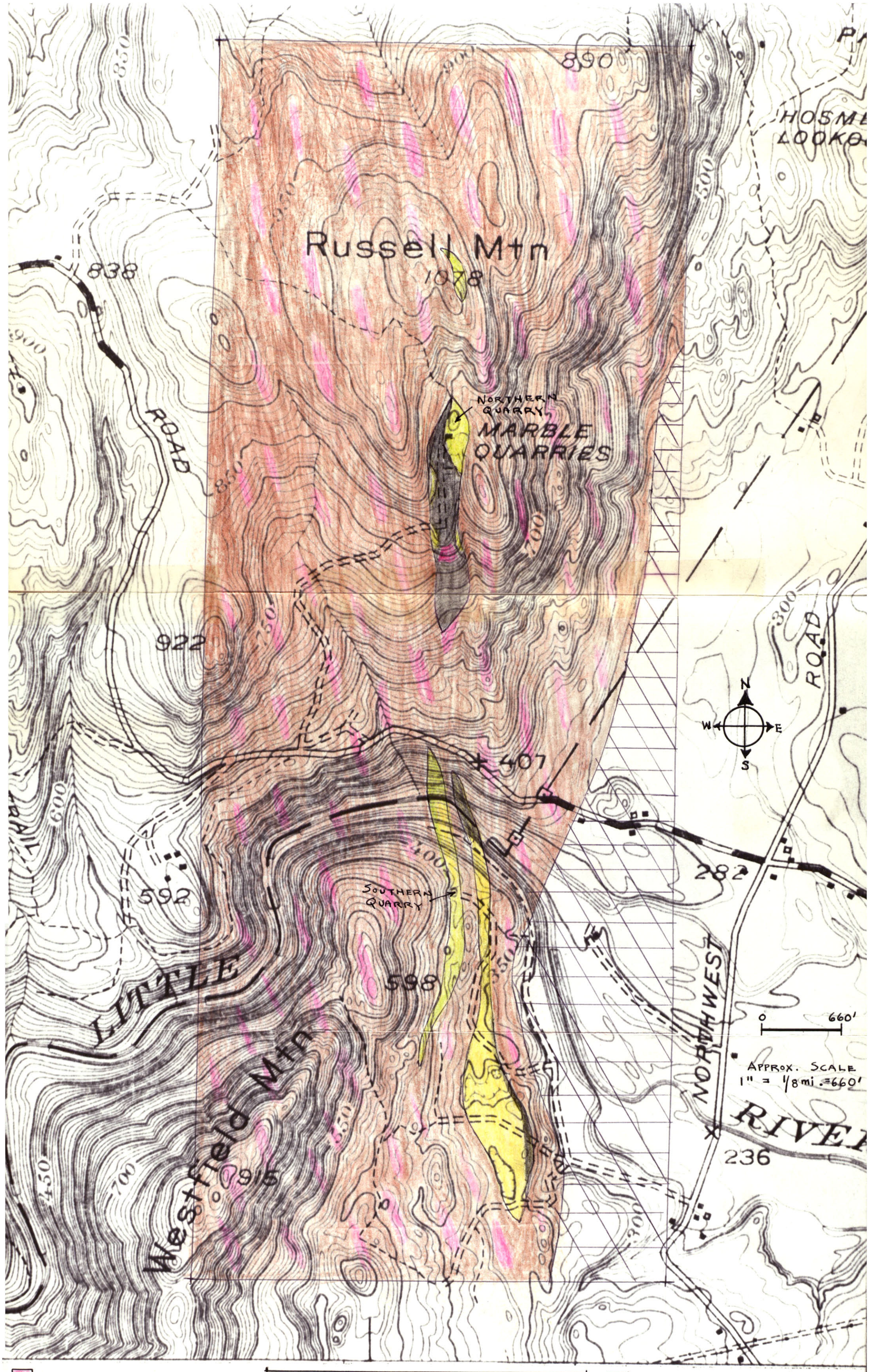
FIELD WORK BY THE AUTHOR

General Geology

Field work was carried out during the fall of 1957 and during the summer and fall of 1958. The area mapped, a rectangle two miles by one mile in size, consists of two abandoned serpentine quarries and the immediately surrounding rocks. This area is located on the western edge of the Connecticut Valley, about eleven miles west of Springfield, and about four miles west of

Westfield. A map of the area on a scale of 1" = 660' is given in Figure 3.

The Little River, a tributary of the Westfield River, cuts through the southern part of the area, forming a steep valley between Russell Mountain to the north and Westfield Mountain to the south. The bottom of the eastern slopes of the two mountains marks the beginning of the lowlands of the Connecticut Valley, whose overburden laps onto these slopes. The general topography of the area reflects the north-south trend of the underlying metamorphic rock. The highest elevation, 1078 feet, occurs at the top of Russell Mountain and the lowest, approximately 240 feet, at the eastern edge of the area. The locality is reached by following Route 20 west from Westfield for approximately four miles and then, turning left, proceeding for about one mile on Northwest Road, thus reaching the intersection of Northwest Road and Western Avenue, which marks the eastern boundary of the mapped area. The northern quarry, located about 1400 feet south of the summit of Russell Mountain, is reached by a trail from Western Avenue, which cuts across the area. A good view of the Connecticut Valley stretching to the east can be obtained from the quarry area. Also to be seen is the Massachusetts Turnpike, which runs east-west approximately 2000 feet north of the mapped area. The southern quarry, located on the northeastern slop of Westfield Mountain near the Little River, is reached by a trail from Northwest Road. A dump on the edge of the Little River to the east of the southern quarry marks the former location



SERPENTINE AREA GEOLOGY
WESTFIELD, MASSACHUSETTS
FIGURE 3.

of a cutting and polishing mill which handled the quarry material. The location and main minerals of these and other quarries and mines in the Springfield vicinity are described by Otis (1952), in a pamphlet available at the Springfield Museum of Natural History.

The only rocks found away from a center line running through the quarries are high-grade pelitic schist and coarse-grained quartz diorite (see Figure 3). Everywhere the quartz diorite intrudes the schist as large separate masses and as small intimate veins and small masses which often appear to grade into the schist. (Although the quartz diorite will be referred to as "intrusive," as used here the term is one of relationship and does not imply any particular mode of origin.) The mountain tops and other high ground are made up mainly of quartz diorite. Along a north-south line determined by the quarries several thin lenses composed of varying combinations of serpentine, amphibolite, and impure marble are intercalated in the schist. These vary in thickness from a few inches to five-hundred feet and in length from six inches to 3/4 mile. They also are intruded by quartz diorite. A large number of serpentine boulders are located on the top and sides of the ridge to the west of the northern quarry but no serpentine outcrops here. The boulders occur in a thin north-south zone for several hundred feet. The schist has a dip (determined from the schistosity) varying from vertical to 60° east or west, and is gently warped, varying in strike from N25E to N25W, except where it wraps around the ends of the larger lenses. The amphibolite and marble are banded and somewhat schistose. The serpentine

is sometimes banded and schistose. The strike of the lenses agrees with that of the enclosing schist. Connecticut Valley overburden encroaches on the southeastern part of the rectangle.

The schist has the texture and mineralogy of a typical high-grade pelitic schist. Porphyroblasts of kyanite and garnet occur in a fine-grained schistose groundmass of biotite, muscovite, oligoclase, quartz, and magnetite. In the field the kyanite and garnet appear to be somewhat mutually antipathetic, with large kyanite grains occurring only when garnet is scarce or absent and vice versa. Sometimes the two are concentrated in separate bands. The schist may contain kyanite and/or garnet, or neither, varying in this respect both across and along the schistosity. The largest kyanite prisms are one inch long and the largest garnets about one-half inch in diameter. Irregular quartz segregations and veins are found throughout the schist. Locally thin stringers of small magnetite grains are found. Quartz diorite veins of varying width parallel and cross-cut the schistosity, with the schist often passing into an irregularly foliated quartz-feldspar gneiss and then into quartz diorite. These quartz diorite masses range from inches up to several hundred feet in diameter. Rarely, distinct schist inclusions may be seen in the quartz diorite. Throughout the area the quartz diorite apparently consists only of quartz, muscovite, and oligoclase, with a rare trace of apatite or beryl.

The lenses, which are restricted to a thin zone about 1000 feet wide, exhibit nearly complete textural and mineralogical gradations between amphibolite, serpentine, and impure marble.

In the vicinity of the northern quarry the amphibolite varies from a banded quartz-actinolitic hornblende-andesine rock through chlorite-and-epidote-bearing rocks to massive serpentine with lesser carbonate and talc. At one contact just north of the Little River, amphibolite is separated from serpentine by a foot of chlorite rock; at another contact, amphibolite grades into six inches of chlorite, then into two feet of talc-actinolite, and finally into massive serpentine-talc rock. The chlorite contacts are sharp and parallel the rock strike. Actinolite "veining" carbonate, massive serpentine, serpentine pods in a talc groundmass, and serpentine pods with thin talc rims in a carbonate groundmass are all seen along the Little River (see Figures 4 and 5). At the southern quarry banded actinolite marble passes directly into banded serpentine marble and into massive serpentine. A similar relationship is found in a lens north of the northern quarry, although this lens is composed mainly of serpentine. Here, the actinolite-carbonate rock is found along the edges of the lens. Diopside-actinolitic hornblende-epidote-calcite-biotite rock is found south of the southern quarry. Elsewhere smaller lenses show various gradations between the quartz-bearing amphibolite and actinolitic hornblende-epidote-andesine amphibolite. The various mineralogical changes and combinations in these rocks, as seen in thin section, will be discussed fully in the next chapter. The relationships of the larger lenses to the schist and pegmatite are shown in Figure 3.

The amphibolite and calc-silicate lenses have sharp contacts with the pelitic schist. The contacts are parallel to

Actinolite "Veining" Carbonate

(scale given by pencil)

Figure 4.

Serpentine Pods (dark green) with Talc Rims (light green)
in a Carbonate Groundmass (brown)

(scale given by pencil)

Figure 5.

the schistosity of the schist and to the banding of the lens rocks. Contacts between marble or serpentine and schist are apparently sharp, although these are seldom exposed and quartz diorite has been intruded at the contact areas which can be seen. With the exception of the schist, the quartz diorite has sharp contacts with all the rock types. Quartz diorite and quartz veins and masses in the amphibolite show no visible contact effects on the intruded rock, but the quartz diorite may become fine-grained within a few inches of the contact. Where the quartz diorite has intruded serpentine or marble, thin monomineralic bands of mica (magnesium-rich biotite; see Chapter V) and actinolite have developed at the contact. These vary from less than an inch to one foot in width, with the actinolite next to the marble or serpentine and the biotite next to the quartz diorite. All contacts are generally sharp. Either or both of the contact minerals may grow perpendicular to a particular contact. At one small area at the southern quarry similar banding is found between schist and serpentine-marble. No other contact effects are visible in the marble, but talc has often developed in the serpentine. Sometimes abundant black tourmaline occurs in the quartz diorite next to the biotite band. At these contacts the quartz diorite is more or less fine-grained adjacent to the biotite band, but grades rapidly to its normal coarse grain within a few inches to a foot. Also, near the contact the quartz diorite has noticeably less quartz and sometimes carries biotite in place of, or with, muscovite.

Northern Quarry Area

Two main types of serpentine rock are seen along the north wall of the northern quarry, which is roughly a square, seventy-five feet on a side. One type consists of small amorphous masses of dark serpentine evenly scattered through a talc groundmass (Figure 6). The other is massive black serpentine with lesser talc, carbonate, and fine-grained, platy green serpentine (Figure 7). In places, the latter type has thin vertical stringers of chrysolite serpentine. Between these is found aphanitic, schistose, greenish actinolitic hornblende-epidote rock with numerous magnetite grains scattered through it. The rest of the serpentine rock in this area is mainly massive black serpentine with varying amounts of talc and carbonate. In the quarried material dumped around the area can be found a little banded actinolitic marble, which is similar to that occurring at the south quarry. Also outcropping in the quarry area, and grading into the serpentine, are a variety of hybrid rocks, apparently representing a transition from amphibolite to serpentine. Their mineralogy consists of various combinations of chlorite, epidote, actinolitic hornblende, and biotite. These hybrid rocks tend to develop sharp contacts with each other. At one spot where this gradation occurs thin serpentine prisms, pseudomorphous after actinolite, are scattered in a talc groundmass (Figure 8).

At the east side of the quarry approximately three feet of talc separate the serpentine from about two feet of banded quartz diorite with intercalated thin biotite bands. Pelitic

North Quarry Wall Exhibiting Small Serpentine Masses
Scattered through Talc Groundmass

(scale given by pick)

Figure 6.

North Quarry Wall Composed of Massive Black Serpentine, Platy Green Serpentine, Light Green Talc, and Brown Carbonate.

Thin vertical chrysotile stringers are seen in lower left-hand corner.

(scale given by pick)

Figure 7.

Thin Serpentine Prisms, Pseudomorphous after Actinolite,
Scattered in Talc Groundmass.

(scale given by knife)

Figure 8.

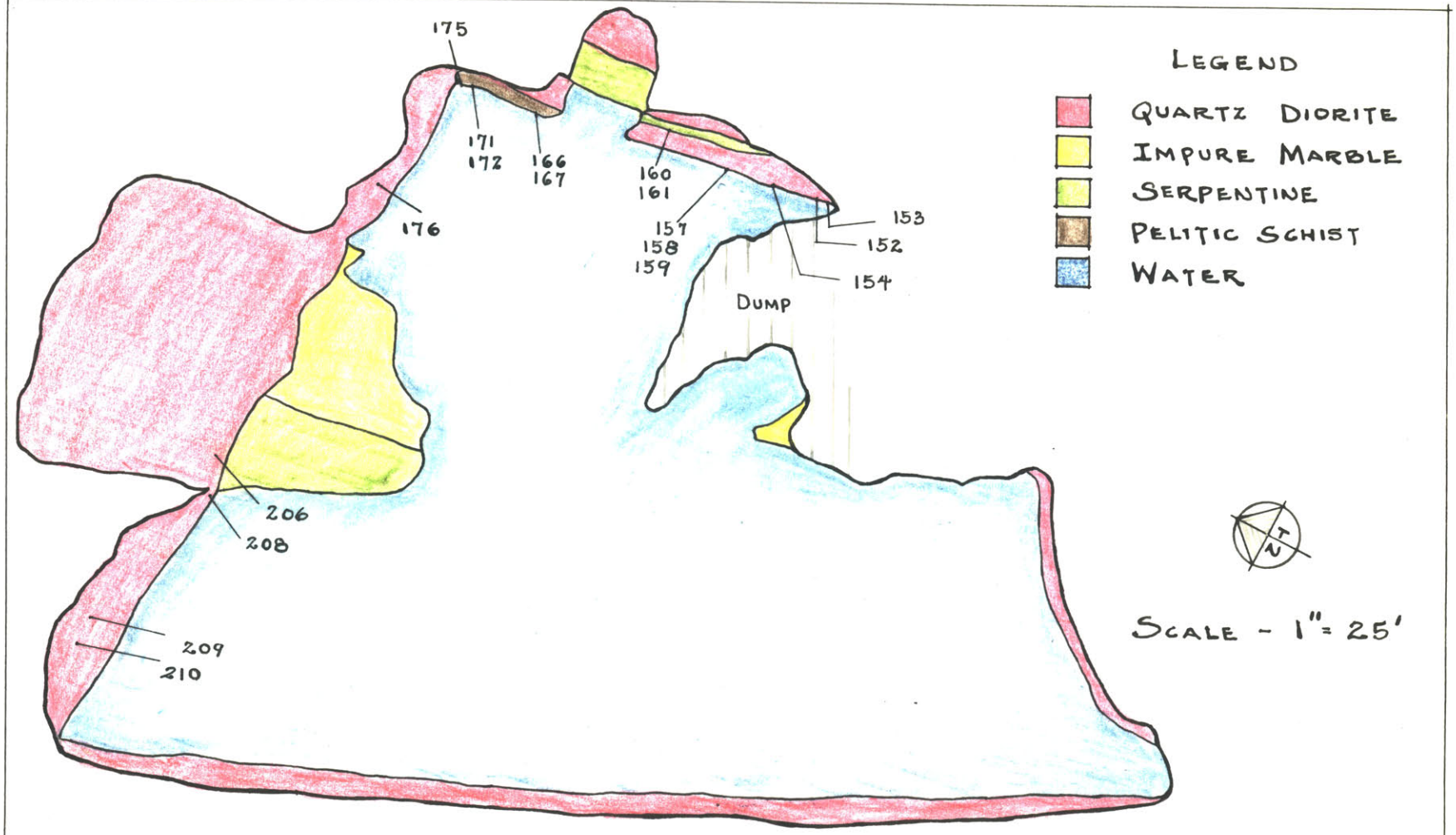
schist is next to the quartz diorite. The banding and schistosity parallel the contact. A contact between quartz diorite and serpentine makes up the south wall of the quarry. Here a one inch biotite band is located next to the quartz diorite, with the biotite growing perpendicular to the contact. The quarrying has removed the serpentine and any other contact minerals which may have been formed. The quartz diorite is fine-grained at the contact, but coarsens to one-inch grains within a foot. At one spot black tourmaline prisms up to three inches long are found with the biotite banding.

The serpentine mass of the quarry area occurs at the northern end of a large amphibolite lens. The lens is approximately 300 feet wide and 2000 feet long. To the south three smaller serpentine bodies are found on the flanks of the amphibolite. These are similar to the massive black serpentine found at the quarry. The side nearest the schist of one of these is mainly an actinolite-talc mixture. The serpentine contacts with the amphibolite and schist are not exposed. The schist-amphibolite contacts are sharp and parallel the banding and schistosity. The dark amphibolite is distinctly banded and is composed of quartz, andesine, and actinolitic hornblende, with lesser biotite and epidote.. Muscovite is sometimes found in the amphibolite. It is often intruded by quartz diorite, with sharp contacts and no sign of contact effects. At both ends of the lens the schist appears to wrap around the amphibolite, as shown by the strike of the schistosity. A large mass of quartz diorite is found at the south end of the lens.

Southern Quarry Area

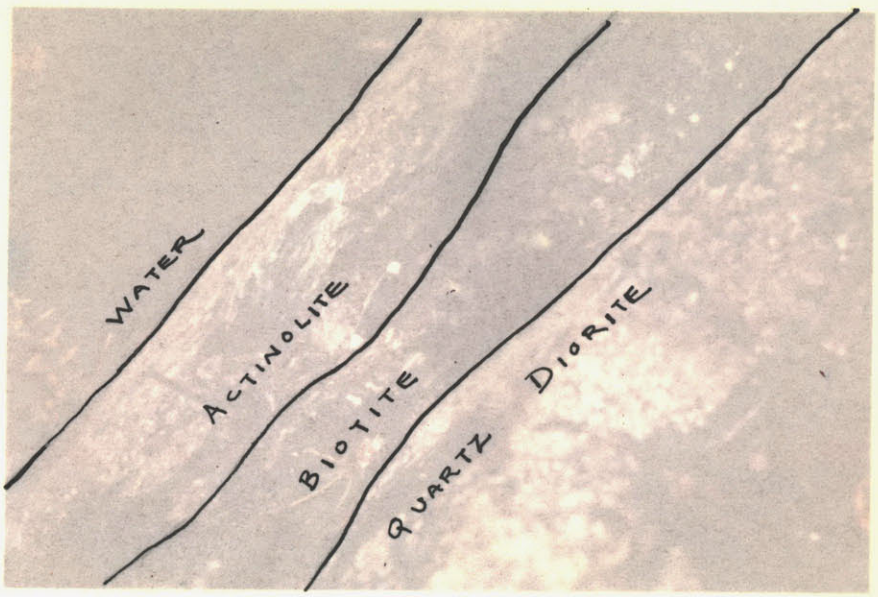
The east, north, and west walls of the southern quarry, partially shown in Figure 9, are located along the former contact between intrusive quartz diorite and marble and/or serpentine. Roughly speaking, the quartz diorite has been intruded parallel to the strike of the marble and serpentine along the east and west walls, and directly across it at the north wall. Part of the serpentine-marble has not been removed and juts out from the middle of the north wall into the quarry. There is little outcrop at the southern end of the quarry, but it appears that the quartz diorite has cut off at least part of the serpentine-marble lens here too.

More or less well developed biotite and actinolite banding is found along the walls. The actinolite band, which is separated from the quartz diorite by the biotite band, is often missing due to the effects of the quarrying. It is best seen along the east wall (Figure 10). At least a trace of the biotite banding can usually be seen on the quartz diorite walls. At some places along the east wall small biotite pods occur in the actinolite band, while at others actinolite pods are in the biotite. Rarely, a small pod of feldspar is seen in the biotite band. The actinolite band is up to a foot wide, and the biotite band up to four inches thick. A few feet from the edge of the east wall, and parallel to it, two thin strips of talc-marble-serpentine occur in the quartz diorite. These exhibit similar banding at their edges. At these edges, and also along the east



LOCATION OF ANALYZED MINERAL SAMPLES
SOUTHERN QUARRY, WESTFIELD, MASS.

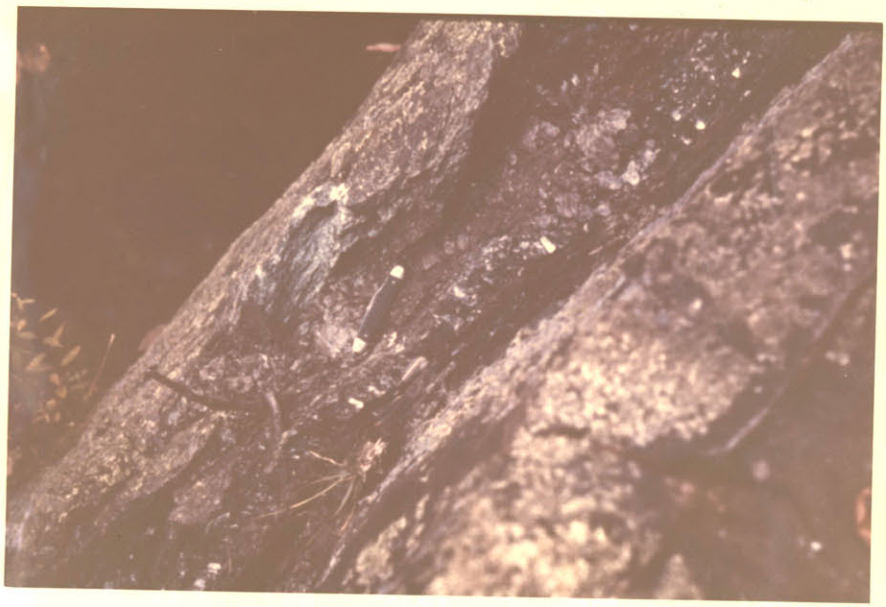
FIGURE 9.



Actinolite and Biotite Bands Along East Wall of Southern Quarry

(scale given by knife)

Figure 10.



Actinolite and Biotite Bands Along East Wall of
Southern Quarry

(scale given by knife)

Figure 10.

wall, banded pelitic schist is sometimes found at the contact with the biotite banding and grades in one or two feet into quartz diorite. The banding is similar next to the schist and next to the quartz diorite. Samples for the mineral analyses reported in Chapter V were taken at these contacts.

The remnant serpentine-carbonate mass adjacent to the north wall is of interest because it shows a change from impure marble into massive serpentine and also the contact relationships of these rocks to the quartz diorite. The eastern half consists mainly of fine-grained, thinly banded actinolitic marble which in places grades into thin, almost pure actinolite rock with scattered fine-grained magnetite. Locally, incipient formation of serpentine in narrow bands can be seen. To the west the marble grades rapidly into massive serpentine. This gradation does not directly coincide with the banding and is apparently not related to original bedding. The serpentine of this western half also shows a local development of actinolite-magnetite rock. The two main rock types probably represent the western and middle beds of the three varieties reported at this quarry by Crosby and Emerson.

Although weathering has altered the contact of this mass with the quartz diorite, there appears to be no major change in the marble or serpentine as the contact is approached. A slight increase of actinolite in the marble occurs a few inches from the contact. It is probable that a very thin actinolite band existed next to the biotite band and graded rapidly into the actinolitic marble or serpentine. The quartz diorite wall has remnants of a

biotite band. The quartz diorite is fine-grained at the contact and rapidly coarsens away. It has biotite instead of muscovite in the contact area, and has abundant black tourmaline and little quartz at the contact. In the next chapter several thin sections of the serpentine-marble mass, including some from the contact area, will be discussed.

About fifty feet to the north of the above contact area there occur a few outcrops of mixed serpentine-marble. Massive serpentine, with some amphibolite, occurs further north in the Little River. To the south of the quarry several small masses of serpentine occur in a thin north-south line. These are separated from the quarry and from each other by quartz diorite, but no contacts are exposed. Further south the serpentine appears to pinch out. About 150 feet east of the line of the serpentine masses and separated from them by pelitic schist, is found a small quarry area where a bed of coarse diopside-actinolitic hornblende-calcite-biotite-epidote rock, approximately seventy-five feet thick, has been opened. Although little outcrop can be found to the north of this bed, it is probably a continuation of a thin amphibolite bed separated by pelitic schist from the serpentine outcropping in the Little River (see Figure 3). Between these two main lenses a few small lenses, measured in inches and a few feet, are scattered through the schist. These smaller lenses are composed mainly of actinolitic hornblende and epidote, with lesser andesine. A sinkhole area with amphibolite outcropping on its eastern edge is encountered south of the small quarry. This is probably a continuation of the bed found at the small quarry.

CHAPTER III. PETROGRAPHY*

PELITIC SCHIST

A study of twenty-five thin sections indicates that the pelitic schist, which makes up most of the area mapped, exhibits little variation in mineralogy. It is a typical high-grade pelitic schist, belonging to the amphibolite metamorphic facies of Turner and Verhoogen (1951), recently renamed the almandine amphibolite facies (Fyfe, Turner & Verhoogen, 1958). The minerals almost always present are biotite, muscovite, quartz, oligoclase, almandine garnet, and kyanite. In addition to these staurolite, sillimanite, and cordierite occur in some of the sections. Usually present are accessory tourmaline and opague.

The parallel biotite and muscovite flakes are intimately associated, with the biotite usually light to dark brownish green in color and containing small zircon inclusions with pleochoric haloes. The biotite sometimes forms a slight replacement of kyanite, feldspar, garnet, and staurolite. Quartz of varying size occurs as independent anhedral masses and as inclusions in garnet, kyanite, staurolite, and feldspar. Oligoclase is found mainly as coarse porphyroblasts with numerous quartz inclusions. There is slight sericitic alteration of the feldspar, which often exhibits good albite and/or pericline twinning. Garnet usually makes up about one per cent of a section, occurring as rounded porphyroblasts scattered among the other minerals. It commonly contains

*(Note: Thin section terms of size used here refer to 10X magnification. Identification of feldspar and carbonate minerals was made by index of refraction measurements.)

numerous inclusions of quartz and opaque. The arrangement of inclusions in some grains indicates rotation during growth of the garnet. Some of the grains are broken. Kyanite appears as large broken plates with ragged edges and many quartz inclusions. Staurolite is found in a few sections as porphyroblasts with quartz and opaque inclusions. Sillimanite occurs in small amounts in several of the sections as clusters of fine-grained fibers and prisms forming from biotite. Cordierite is seen rarely as a few anhedral grains with polysynthetic twinning. Accessory grey-green tourmaline is scattered through the sections as rounded grains and thin plates. Anhedral masses and thin plates of opaque, probably representing magnetite and ilmenite respectively, make up about two per cent of each section.

QUARTZ DIORITE

The intrusive quartz diorite is composed of approximately equal amounts of quartz and oligoclase, with lesser muscovite and, rarely, with a trace of apatite or beryl. Tourmaline and biotite occur in contact areas. A very coarse-grained aplitic texture is displayed in thin section. The feldspar grains are homogeneous and almost all exhibit albite twinning. Numerous quartz inclusions occur in the feldspar, and the two minerals are often intimately intergrown. No potash feldspar was found in the relatively few thin sections studied.

CALCAREOUS ROCKS

In this group are combined the amphibolite, serpentine, and marble, and the various gradations between them. In general no

clear-cut break between these three rock types is found in the field; a similar relationship exists in thin section. The north-south lenses in which these rocks are found become more calcareous toward the south, and will be treated here in terms of location, starting at the northern quarry and moving south. This discussion is based on a total of forty-four thin sections.

Most of the amphibolite lens running south from the northern quarry is composed of andesine, quartz, and actinolitic hornblende. Minor (less than five per cent) minerals found are biotite, epidote, and opaque, as well as a trace of sphene. Near intrusions of quartz diorite, chlorite becomes abundant at the expense of quartz, with the chlorite appearing to form from the feldspar and containing inclusions of it and opaque. The actinolitic hornblende occurs as fine-grained to very coarse-grained ragged porphyroblasts with numerous inclusions of quartz, feldspar, and opaque. It is pleochroic from light green to dark blue-green. Quartz is found as rounded masses interlocking with feldspar to form the groundmass of the sections. The feldspar sometimes has quartz inclusions, and shows varying degrees of sericitic alteration. Some albite and lesser pericline twinning is seen. Opaques make up one to five per cent of each section as scattered rounded plates and masses. Colorless epidote, when found, tends to be segregated in parts of a section as rounded grains with opague inclusions and anomalous blue interference colors. Biotite also occurs locally as a replacement of the hornblende. A small amount of sphene makes up aggregates of rounded grains. The texture of

these sections may show parallel orientation of the hornblende, depending on the relationship of the individual sections to the rock schistosity and lineation. In the field thin banding of light and dark minerals is often seen.

In the vicinity of the northern quarry, and grading into serpentine, are found small amounts of a number of hybrid rocks, apparently representing a transition from the amphibolite to the serpentine. Assemblages found include actinolitic hornblende-epidote, actinolitic hornblende-epidote-chlorite, actinolitic hornblende-epidote-opaque, and biotite-brucite. The brucite of these rocks, and also that reported with serpentine, is characterized by a reddish-brown interference color, and actually may be another chlorite mineral. On a small scale, strong segregation of any one of these minerals may be found. Examination of more thin sections would probably disclose other assemblages. Accessories in these sections, only one or two of which occur in a given section, include plagioclase, feldspar, opaque, antigorite, biotite, brucite, apatite, carbonate, spinel, and sphene.

The actinolitic hornblende is fine-grained to coarse-grained, anhedral, pleochroic from light green to light to dark blue-green, and usually has inclusions of epidote. Epidote is colorless to light green, makes up the groundmass of a section, and has anomalous blue interference colors. Green chlorite is strongly pleochroic, anhedral, and is found as rounded to thin elongated masses. Opaques occur as irregular masses with ragged edges. They tend to be associated with biotite and may have inclusions

of many of the other minerals. In one section the opaques are surrounded by rims of sphene. Scattered ragged shreds and flakes of biotite are green to brown in color and slightly pleochroic. The biotite may have inclusions of opaque, epidote, and hornblende. Brucite makes up fine-grained to coarse-grained fibrous plates and is often intimately associated with biotite. In one section very fine-grained antigorite flakes grade into brucite. Apatite forms rounded grains some of which are included in biotite. Accessory carbonate is found as scattered anhedral masses. Dark green spinel is associated with biotite and has opaque inclusions. Plagioclase feldspar seems to be forming from epidote in one section. The texture of these sections varies between those with preferred orientation of the major minerals and a random orientation.

Thin sections of the northern quarry serpentine mass are made up of varying amounts of antigorite, carbonate, and talc, with lesser opaque and brucite. One per cent or less of a chlorite mineral with an "ultrablue" interference color is often found. Antigorite usually occurs as a groundmass of aphanitic to fine-grained, flaky material with random orientation. Locally, larger flakes with a higher birefringence grade from this material. Often thin, discontinuous veins of slightly higher birefringent serpentine with opaque center-lines cut the main mass. The vein serpentine grows perpendicular to the vein walls. Very fine-grained to medium-grained scattered masses and flakes of opaque are intimately mixed with the antigorite. In contrast to this

texture, which gives no direct clue to the mineral or minerals from which the serpentine formed, one or more pseudomorphs of antigorite after coarse-grained forsterite or of antigorite after fine-grained to coarse-grained actinolite are found rarely. The actinolite pseudomorphs occur as thin prisms, while the almost completely replaced forsterite is found as rounded dark patches. The alteration of these two minerals to serpentine is not as advanced in sections from the southern quarry. More details of this replacement will be given in the discussion of those sections. (See Figures 11, 12, and 13.)

Irregular masses of carbonate of all sizes occur throughout some sections, while only trace amounts are found in others. Index measurements indicate most of the carbonate to be dolomite. It sometimes occurs as thin, discontinuous veins and may be intimately mixed with the other minerals. Wavy flakes and fibers of talc are scattered through the antigorite. In some sections the talc appears to be replacing the antigorite, in others the reverse holds true. In most, no definite relationship is seen. Fine-grained to coarse-grained brucite sheaves and flakes are scattered through the antigorite, usually in association with opaque. The brucite has a reddish-brown interference color and individual sheaves are often surrounded by a rim of ultrablue chlorite. Opaques make up as much as fifteen per cent of a section, occurring principally as very small masses and stringers disseminated in the antigorite. Larger masses are associated with brucite, talc, carbonate, and ultrablue chlorite. In one

section two sets of parallel trains of opaques, tending to run perpendicular to each other, form a network throughout the section. Flaky ultrablue chlorite is associated with carbonate, brucite, talc, and opaque. In places it appears to be forming from antigorite.

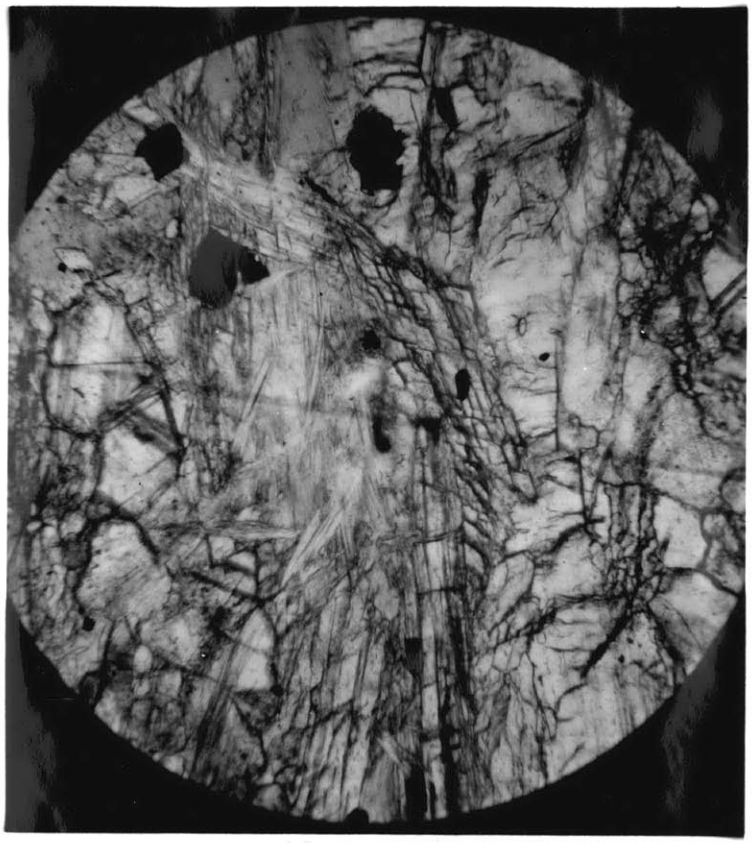
South of the Little River the amphibolite is similar in texture and mineralogy, except that quartz becomes a minor constituent or is not found at all and epidote is now found in abundance. The epidote is colorless to light yellow-green, makes up fine-grained to coarse-grained rounded masses and prisms, and is often included in the actinolitic hornblende.

In a similar southern gradation the serpentine rocks tend to contain more carbonate, and at the southern quarry mixed marble and serpentine is found. Where serpentine is not found in the marble it consists of varying amounts of carbonate, mainly dolomite, and tremolite-actinolite. Crosby (1896) reported that an analysis of the marble found here indicated it to be a nearly pure dolomite. The rounded masses and ragged prisms of amphibole are of all sizes, anhedral, and scattered through the carbonate groundmass with a roughly parallel orientation. About two per cent opaques are also found throughout each section, tending to concentrate in irregular stringers parallel to the oriented amphibole prisms.

Sections from rocks showing a gradation between the marble and massive serpentine exhibit all degrees of replacement of tremolite-actinolite by antigorite, often in association with carbonate. In addition, medium-grained to coarse-grained masses

of forsterite show strong replacement by these minerals. Replacement of the amphibole occurs mainly as thin, irregular veins of antigorite or of mixed antigorite-carbonate. These tend to follow the amphibole cleavage, with the strongest alteration at the edges of the grains (Figure 11). In advanced alteration the amphibole is broken up into low birefringent cleavage fragments "floating" in a "sea" of antigorite and carbonate. With further alteration irregular masses showing only traces of amphibole texture are left. Usually the extinction angle and interference figure are still distinctive.

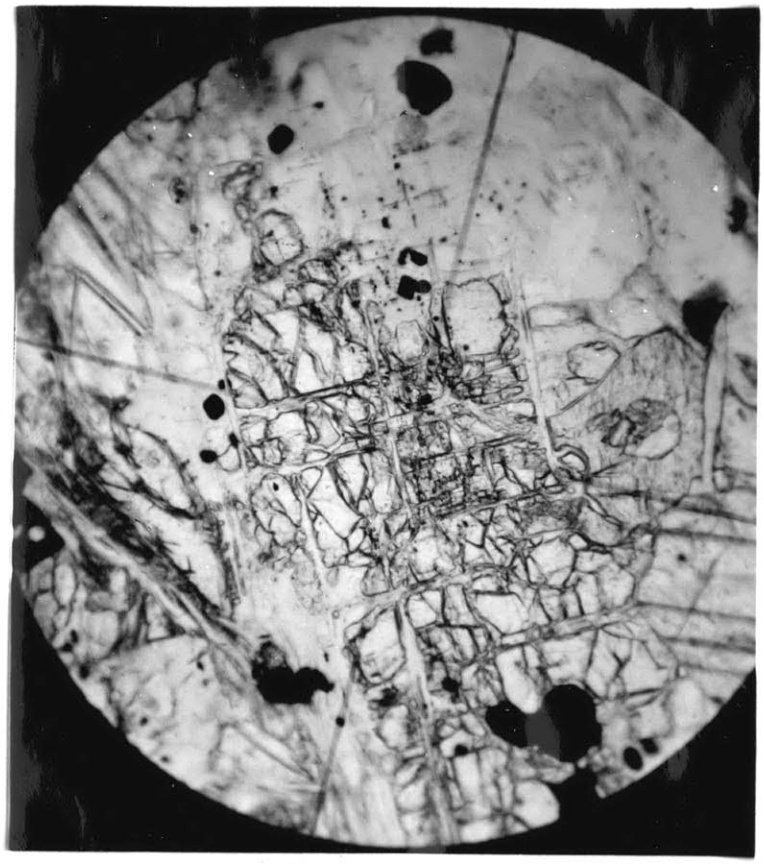
The forsterite is less abundant but appears to have originally made up about half of some sections. It is always more altered than the amphibole, occurring as scattered rounded masses in a groundmass of antigorite and carbonate. The replacement tends to occur along two planes at ninety degrees to each other, with the greatest replacement at the edges of the grains (Figure 12). The resulting antigorite has a mesh texture, often mixed with carbonate. Very fine-grained opaque inclusions may sometimes be seen at the rims of the forsterite grains. In places opaque inclusions in the altering forsterite reflect carbonate cleavage (Figure 13). In a few sections dolomite shows replacement by antigorite, mainly along cleavage and twinning planes (Figure 14). Also the replacement may occur in thin, irregular zones parallel to the section lineation, or as radiating clusters scattered in the carbonate (Figure 15).



Tremolite-Actinolite Grains Being Replaced by
Anhedral Antigorite and Carbonate

(approx. 80X)

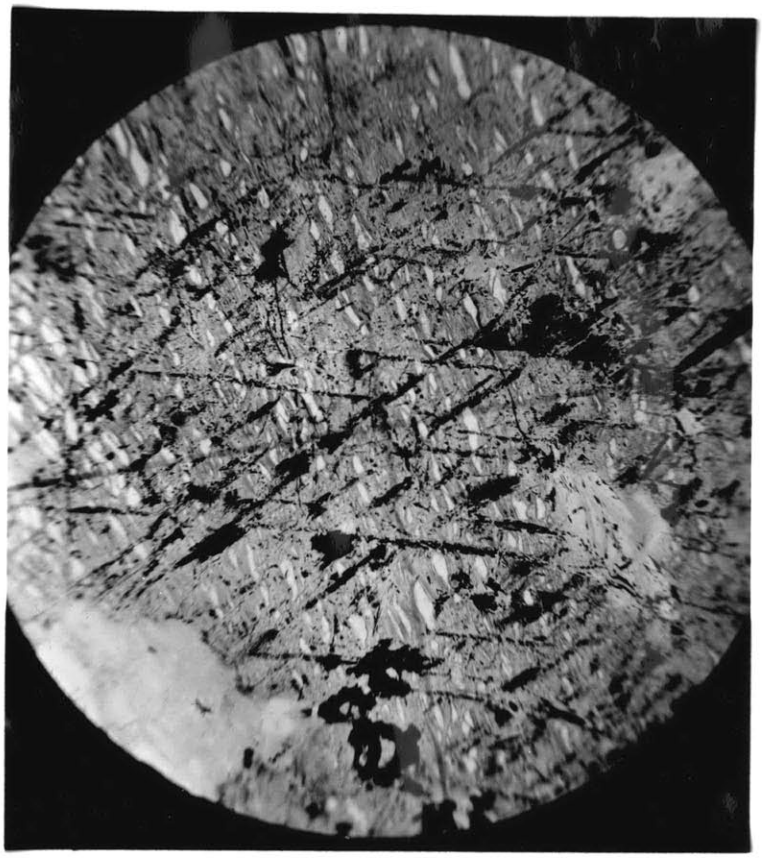
Figure 11.



Forsterite Grain in Advanced Stage of Replacement by Antigorite

(approx. 80X)

Figure 12.

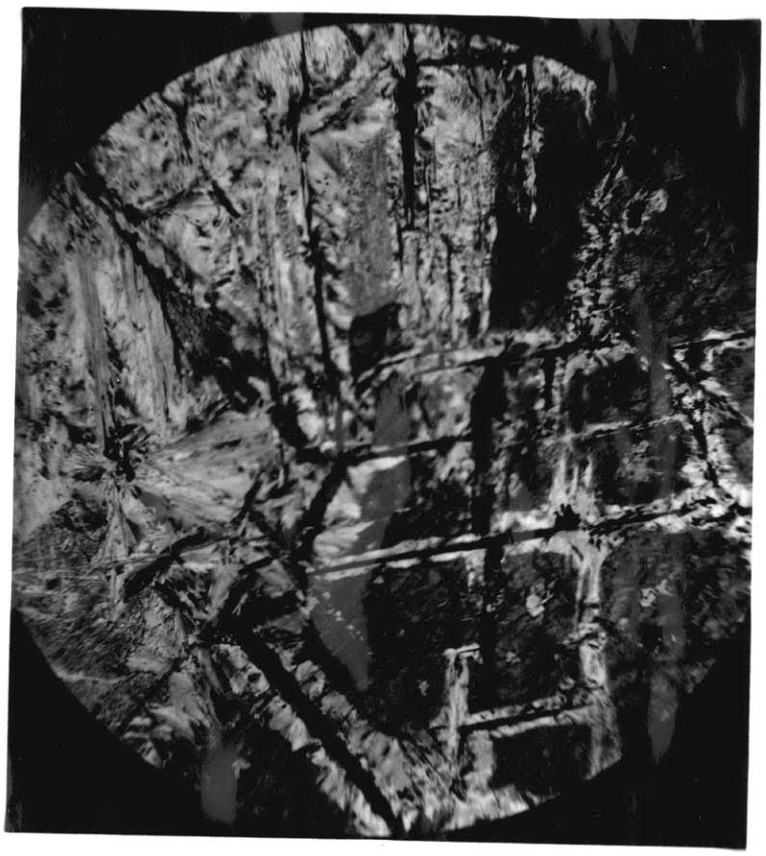


Forsterite (white) Almost Completely Replaced
by Antigorite (light grey)

Opaque inclusions reflect carbonate cleavage.

(approx. 80X)

Figure 13.

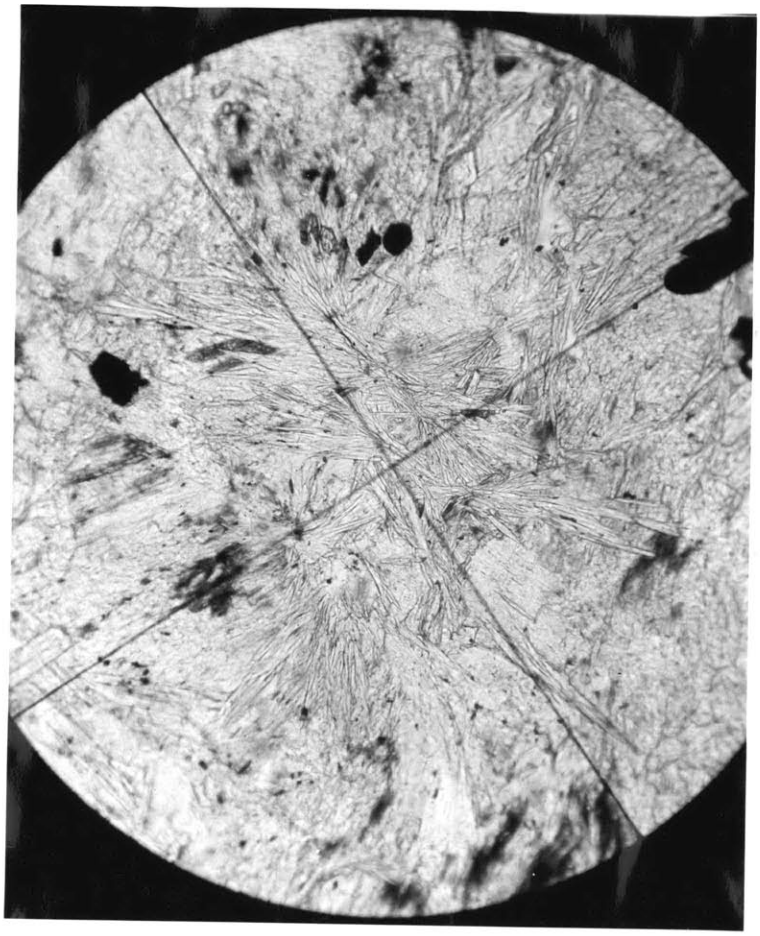


Dolomite (dark grey) Being Replaced
by Antigorite (light grey)

Opaque (black) is associated with the alteration.

(approx. 80X)

Figure 14.



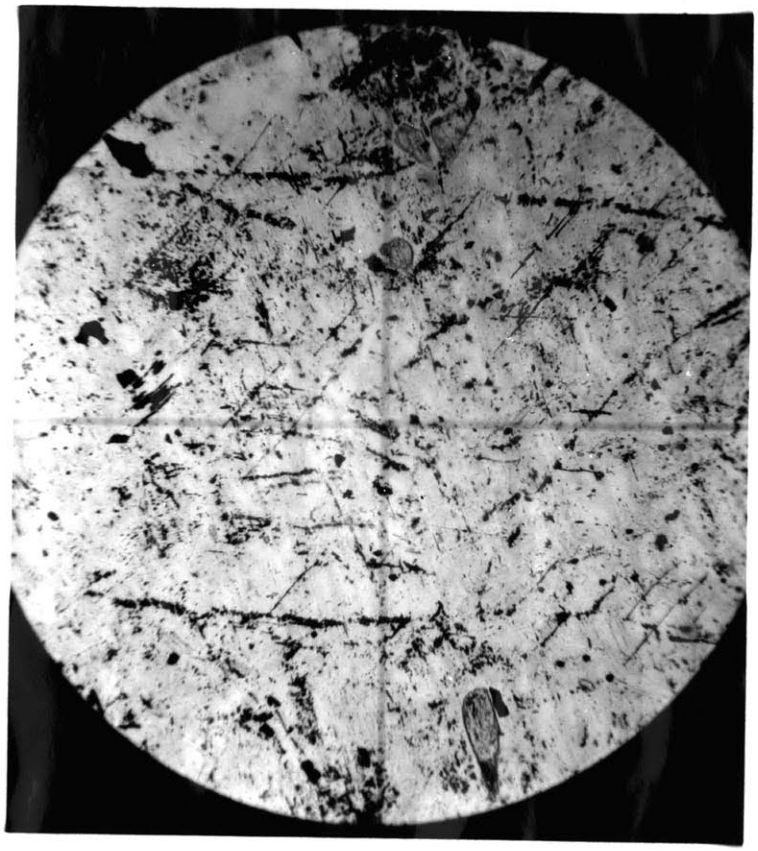
Radiating Clusters of Antigorite Prisms Forming as a Replacement of Dolomite

(approx. 80X)

Figure 15.

These rocks grade into massive serpentine composed only of mesh structure antigorite and a few per cent of opaque. The small opaque inclusions in the antigorite tend to reflect carbonate cleavage (Figure 16). Often two sets of parallel, very thin, irregular veins of ultrablue chlorite, growing in the groundmass and associated with opaque, reflect carbonate cleavage or run perpendicular to each other. Traces of brucite flakes with ultrablue chlorite borders are found in most sections. In places the brucite grades into antigorite.

Calc-silicate rock, including very coarse diopside crystals, outcrops to the south of the southern quarry. Thin sections of this rock have a banded appearance, with some segregation of individual minerals, and consist of mixtures of two or three of the following five minerals: diopside, calcite, biotite, epidote, and gradations between tremolite and actinolitic hornblende, often with minor amounts of opaques. Assemblages found in a fifty-foot section perpendicular to the strike included calcite-amphibole, biotite-amphibole, epidote-amphibole, epidote-amphibole-biotite, calcite-amphibole-biotite, and diopside-amphibole-calcite. The amphibole varies from a colorless tremolite to an actinolitic hornblende similar to that described above, and is pleochroic from light green to light blue-green in the latter case. The diopside is very coarse-grained and exhibits extensive replacement by amphibole and dolomite. The carbonate is found as interlocking grains and as irregular stringers and masses. Biotite is pleochroic from colorless to light green or from light to dark



Opaque Inclusions in Antigorite Reflecting Carbonate Cleavage

(approx. 80X)

Figure 16.

brownish-green, and occurs in segregated groups. It sometimes forms an alteration of amphibole. Epidote occurs as scattered grains which are slightly pleochroic from colorless to light yellow-green. Anhedral opaque masses are usually associated with amphibole and biotite concentrations. A little sphene is found in one section.

It is interesting to note the similarity between the serpentine thin sections described here and those described by Emerson (1898, p. 96) from the Westfield and other serpentine bodies in western Massachusetts. In his "Plate 2" he shows olivine changing to tremolite and tremolite to serpentine at Blandford; dolomite changing to serpentine at Granville; and enstatite changing to serpentine at Granville. He felt that the serpentine structure at Westfield indicated a derivation from enstatite. However the author found no enstatite, nor indication of serpentine derivation from enstatite. Diopside is found south of the southern quarry, although not in the exposed serpentine rocks, and this may have been the mineral identified by Emerson as enstatite. It is possible that diopside also formed in the forsterite bearing rocks and was later replaced by serpentine along with the forsterite.

CONTACT SECTIONS

A number of sections from the contact of quartz diorite or pelitic schist with serpentine and/or marble, marked by biotite and actinolite banding, were studied. At the contact between the quartz diorite and the thin biotite bands an irregular junction

between plagioclase feldspar and mixed tourmaline-biotite is usually found, although a distinct tourmaline zone may separate the biotite from the feldspar. Little or no quartz occurs with the feldspar at the contact, but there may be a few scattered flakes of biotite. Within a foot of the contact the quartz diorite shows its usual texture and association of quartz, plagioclase feldspar, and muscovite. The feldspar often exhibits alteration to very fine-grained chlorite near the contact. This alteration takes the form of scattered masses and very thin, irregular veins, with the veins running perpendicular to the contact from near or at the contact into the feldspar. The chlorite veins show cross-fiber structure, have an anomalous blue interference color, and may also occur in the biotite band. Sericitic alteration of the feldspar also increases toward the contact. In many sections the feldspar is altering directly to biotite and grades rapidly into almost pure biotite. The tourmaline is of all sizes. It may be colorless or pleochroic from light to dark slate blue. The biotite is slightly pleochroic from colorless to very light green, making up irregular plates and masses. Scattered grains and masses of apatite occur rarely in the biotite band.

Near the contact of the pelitic schist with the serpentine rocks the schist displays its regular mineralogy. At the one exposed contact kyanite is not present. Also a small amount of actinolite replaces the biotite in the schist here, as shown by remnant pleochroic haloes from the mica. A one-quarter inch tourmaline band separates the schist from the biotite band.

Two sections of the actinolite band at the southern quarry were studied. In one, prisms and grains of tremolite-actinolite are intimately mixed with carbonate and lesser antigorite, with the amphibole showing all degrees of alteration to these two minerals. The tremolite-actinolite exhibits a general lineation and is slightly pleochroic from colorless to very light green. A trace of the biotite is sometimes intergrown with the antigorite, and the latter has numerous small inclusions of opaque. The opaque is black to reddish-brown. The second section is composed of interlocking actinolite, with a band of clinozoisite running through the center of the section. The clinozoisite band has masses of actinolite which appear to be replacing the clinozoisite. The latter is colorless and has conspicuous ultrablue interference colors. The actinolite is pleochroic from light green to light blue-green. A trace of antigorite occurs as an alteration of the clinozoisite. About five per cent of the section is composed of biotite, which is being altered to actinolite. Also scattered through the section are traces of opaque and apatite.

Within a few feet of the contact of serpentine with siliceous rock at the northern quarry an increase in the amount of talc present appears to be the only change in the magnesian rock. This increase may be slight or may result in the formation of almost pure talc. At the southern quarry the contact effects in the mixed marble-serpentine rocks are limited to a few inches from the cross-cutting quartz diorite. As the intrusive is approached there is a rapid increase in the amount of tremolite-

actinolite present, resulting in almost pure amphibole next to the biotite band at the contact. This contact relationship appears to hold true for the marble with little or no serpentine and with abundant amphibole, and for the almost pure serpentine rock with no amphibole, as well as for the gradations between them.

Two sections from the contact between pelitic schist and one of the small enclosed amphibolite lenses indicate a rapid gradation between the two. This gradation occurs in a few inches and is marked mainly by a strong increase in biotite and decrease in actinolitic hornblende proceeding from the amphibolite to the schist.

ORIGIN OF THE CALCAREOUS ROCKS

The mineralogy and replacement relationships of the calcareous rocks are best explained as due to regional metamorphism of dolomite rock with varying amount and type of original impurity. The sequence of events to be postulated here will be outlined by treating the mineralogical changes brought about by the metamorphism of three different dolomites, each with a given amount and type of impurity at the beginning of metamorphism. The rocks described above probably represent various gradations or combinations of these three cases. Also they would depend on the extent of the reactions and appear to reflect several of these. It should be noted that the order of the reactions is in terms of falling temperature, and most of the changes are thus of a retrograde nature. The effect of the quartz diorite is considered to

be limited to the contact banding except for possible introduction of water.

The first case is that of dolomite with limited silica impurity. The following steps could take place in the order given during regional metamorphism:

1) formation of forsterite marble with a slight loss of volume: $2CaMg(CO_3)_2 + SiO_2 \rightarrow Mg_2SiO_4 + 2CaCO_3 + 2CO_2\uparrow$

Dolomite Quartz Forsterite Calcite

2) as temperature falls, addition of water from an intrusive or by squeezing of surrounding pelitic sediments, forming serpentine by equal volume replacement of forsterite, with magnesia and silica going into solution:

$$5Mg_2SiO_4 + 4H_2O \rightarrow 2Mg_3Si_2O_5(OH)_4 + 4MgO + SiO_2$$

Forsterite Serpentine

Unless a great volume of water is available, solutions resulting from this reaction will be greatly concentrated in magnesia and silica.

3) solutions from above react with dolomite to form tremolite and serpentine, generally parallel to imposed metamorphic structure, but locally also cross-cutting the structure:

$$2CaMg(CO_3)_2 + 3MgO + 8SiO_2 + H_2O \rightarrow Ca_2Mg_5Si_8O_{22}(OH)_2 + 4CO_2\uparrow$$

Dolomite Tremolite

$$CaMg(CO_3)_2 + 2MgO + 2SiO_2 + 2H_2O \rightarrow Mg_3Si_2O_5(OH)_4 + CaCO_3 + CO_2\uparrow$$

Dolomite Serpentine Calcite

4) as temperature continues to fall, excess magnesia plus carbon dioxide and water react with tremolite to form serpentine and calcite:

$$Ca_2Mg_5Si_8O_{22}(OH)_2 + 7MgO + 7H_2O + 2CO_2 \rightarrow 2CaCO_3 + 4Mg_3Si_2O_5(OH)_4$$

Tremolite — Calcite — Serpentine

5) at these lower temperatures talc may form by addition of silica from an intrusive or by reaction of serpentine with carbon dioxide:

$$Mg_3Si_2O_5(OH)_4 + 2SiO_2 \rightarrow Mg_3Si_4O_{10}(OH)_2 + H_2O$$

Serpentine — Talc

$$Mg_3Si_2O_5(OH)_4 + 3CO_2 \rightarrow Mg_3Si_4O_{10}(OH)_2 + 3MgCO_3 + 3H_2O$$

Serpentine — Talc — Magnesite

The second case to be discussed is that of dolomite with abundant silica impurity. The above series of reactions would be altered in the following way:

1) because of the greater abundance of silica, diopside would form instead of forsterite:

$$CaMg(CO_3)_2 + 2SiO_2 \rightarrow CaMgSi_2O_6 + 2CO_2 \uparrow$$

Dolomite — Quartz — Diopside

2) at lower temperatures and with the addition of water, formation of tremolite from diopside and dolomite, or from dolomite and silica:

$$4CaMgSi_2O_6 + CaMg(CO_3)_2 + CO_2 + H_2O \rightarrow Ca_2Mg_5Si_8O_{22}(OH)_2 + 3CaCO_3$$

Diopside Dolomite Tremolite Calcite

$$5CaMg(CO_3)_2 + 8SiO_2 + H_2O \rightarrow Ca_2Mg_5Si_8O_{22}(OH)_2 + 3CaO_3 + 7CO_2 \uparrow$$

Dolomite Quartz Tremolite Calcite

There would be no serpentine formed since forsterite would not be present for alteration. Thus, too, there would be no opportunity to form excess magnesia for further development of serpentine as in the first case. Also the main carbonate mineral in these rocks, if any, would be calcite, since dolomite would be used up due to excess silica.

In the third case, where a dolomite contains both silica and argillaceous impurity, several new minerals may be formed by regional metamorphism. The resulting rocks consist of various combinations of these new minerals, together with some of those of the first two cases, depending on the relative proportions of silica, lime, magnesia, and alumina in the original sediment and on the degree of metamorphism. The presence of iron as an impurity may become important here also. In the case of very impure dolomite, minerals which can be found in medium-grade rocks of this type include biotite, actinolitic hornblende, epidote, plagioclase, and diopside. The amphibolite and calc-silicate rocks found at Westfield are most likely of this origin.

CHAPTER IV. SOME ASPECTS OF MINERAL EQUILIBRIUM

DEVELOPMENT AND APPLICATION OF TWO GEOLOGIC PHASE RULES

The object of a phase rule is to state how many things must be known about a system in order to predict all of its other properties and characteristics. In the field of geology we are primarily interested in the values of the intensive variables (not dependent on total mass) which characterize the system, i.e., pressure, temperature, and molar fractions of all c components in all p phases. A phase rule is obtained simply by counting the number of unknowns and the number of relations necessary to determine them.

Before developing any phase rules it is pertinent to consider two thermodynamic properties of any system. The Gibbs free energy of a particular system is a property characteristic of that system at any given pressure and temperature. The chemical potential μ of a component i in a system is defined as the change in Gibbs free energy per change of mole of component i at constant temperature, constant pressure, and constant number of moles of the other components. Mathematically this is expressed as: $\mu = \left(\frac{\partial G}{\partial n_i}\right)_{T, P, n_j, n_K, \text{etc.}}$

The chemical potential of a component in a phase can be considered as a measure of the potential of the component to change from that phase to another. A necessary condition for a component to migrate spontaneously from phase a to phase b is that its chemical potential be greater in phase a than in phase b. For

instance, water will vaporize only if its chemical potential is greater in the liquid phase than in the vapor phase:

$$\mu^{\ell}_{H_2O} > \mu^{v}_{H_2O} .$$

When the two phases are in equilibrium no transfer occurs, and:

$$\mu^{\ell}_{H_2O} = \mu^{v}_{H_2O} .$$

A necessary condition for equilibrium in a system of several components and several phases is that the chemical potential of each component be the same in all phases in which it is present.

Two geologic phase rules can now be developed. Consider a heterogeneous system of c components in p phases. Equilibrium requires that the chemical potential of each component should be the same in all phases in which it is present, so that we have $c(p-1)$ equations stating that the system is in equilibrium:

$$\begin{array}{ccccccccccccc} \mu_1^1 & = & \mu_1^2 & = & " & " & " & " & " & " & " & " & \mu_1^p \\ " & & " & & & & & & & & & & " \\ " & & " & & & & & & & & & & " \\ \mu_c^1 & = & \mu_c^2 & = & " & " & " & " & " & " & " & " & \mu_c^p \end{array}$$

Now the chemical potentials depend on pressure, on temperature, and on the molar fraction of each component in every phase; there are thus altogether $2 + cp$ variables P, T, $\bar{n}_1^1, \bar{n}_1^2, \ldots \bar{n}_1^p, \ldots \bar{n}_c^p$. All the molar fractions are not mutually independent, since for each phase $\sum_i \bar{n}_i = 1$; so that there are p additional relations of this type. All told, there are $c(p-1) + p$ relations between $cp + 2$ variables. Subtracting the relations from the variables gives the number of arbitrary values which must be assigned to determine all the other variables from these arbitrary values. This quantity, f, is the number of degrees of freedom of

the system, and the relation $f = c + 2 - p$ expresses the Gibbs phase rule. It states that the state of any system is defined, as far as the intensive variables P, T, $\bar{n}_1^1$, $\bar{n}_1^2$, ... $\bar{n}_1^p$, ... $\bar{n}_c^p$ are concerned, by f of these quantities. If any component i is not present in any phase a, there is no equilibrium condition involving μ_i^a, and therefore there is one equation less; but there is then the additional relation $\bar{n}_i^a = 0$, and the number of degrees of freedom remains unchanged.

Consider a one component system consisting of Al_2SiO_5. If the phases present in this system are kyanite, andalusite, and sillimanite, then $f = c + 2 - p = 1 + 2 - 3 = 0$. This means that the three phases, kyanite, andalusite, and sillimanite, can coexist at only one value of the temperature and pressure. If either the temperature or pressure is varied, then the conditions for equilibrium are no longer satisfied. Because pressure and temperature vary from place to place in the earth the chance of finding a geologic system with zero degrees of freedom is very remote. Such a system could exist in equilibrium only at one particular spot where the temperature and pressure would be just right. Actual mineral assemblages appear to remain stable under variable conditions of pressure and temperature, so that their degrees of freedom, or varience, must be at least two. But if $f \geq 2$, $p \leq c$, so that the number of phases (minerals) will be at most equal to the number of components. This is Goldschmidt's mineralogical phase rule. (Turner and Verhoogen; 1951, p. 423.)

A special case of this phase rule occurs when a fluid phase is present. This introduces a new variable, since the pressure on the fluid may be different from that on the solid phases. Examples would be the pressure of liquid water or carbon dioxide gas in a system which has a solid under a different pressure. The phase rule is then given as $f = c + 3 - p$, with the degrees of freedom considered to be at least three in the development of Goldschmidt's mineralogical phase rule.

The assumptions of the Goldschmidt phase rule are: 1) no fluid phase present; 2) temperature and pressure are the only variable external factors and are constant throughout the system; 3) no mobile (moving) components; or, alternatively, 1) fluid phase present with different pressure than that on solid phases; 2) temperature, solid pressure, and fluid pressure are the only variable external factors and are constant throughout the system; 3) no mobile components. The case of moving components will be considered shortly.

The decision as to what should be considered a "component" depends on the case in question. In general, the number of components c should be the minimum number of different chemical species required to describe adequately the chemical composition of the system. In most cases the simple oxides, such as SiO_2, CaO, MgO, etc., will serve the purpose. In other cases more complex molecules may be used. Any choice of components is justified from the point of view of the phase rule as long as the composition of all phases concerned may be expressed in terms of the chosen

components, and provided, of course, that their number is minimal for this purpose.

We can now consider two distinct cases, or means of application, of Goldschmidt's mineralogical phase rule. The first is that of a closed system. A system is said to be closed if no masses are added or subtracted from it in the course of the transformation under consideration. For this case we can use the general form: $f = c + 2 - p$. Assuming at least two degrees of freedom (pressure and temperature) results in the number of minerals (phases) equalling the number of components. Consider for example an outcrop of banded marble containing calcite, dolomite, forsterite, actinolite, serpentine, talc, and magnetite. The components are CaO, MgO, FeO, SiO_2, Fe_2O_3, CO_2, and H_2O. Thus, minerals equal components, and this assemblage could have formed in a closed system with equilibrium attained as metamorphism proceeded. The forsterite, actinolite, serpentine, talc, and magnetite would be metamorphic minerals derived from an original impure dolomitic limestone. We could vary a CO_2 - H_2O gas pressure as well as the solid pressure, giving one more degree of freedom. In this case the phase rule is given as $f = c + 3 - p$, and a similar result is found by assuming the two pressures and the temperature to vary.

Now consider a quartz vein in a typical pelitic schist. Assume that the minerals composing the schist are quartz, muscovite, biotite, kyanite, almandine garnet, and staurolite. Thus the independent components of the schist (system) are SiO_2, K_2O, Al_2O_3,

H_2O, FeO, and MgO -- a total of six. When the schist was undergoing metamorphism the six minerals, plus a fluid phase, can be considered to have been present. The phase rule for a closed system is then: $f = c + 3 - p$. Assuming at least two degrees of freedom gives: phases equal components plus one. The fluid phase can be postulated to disappear at the end of crystallization, thus giving: components equal phases. Therefore the quartz vein could have formed without introduction of material, since there are six components and six minerals. If any other minerals were present in the schist in an appreciable amount, this would indicate that equilibrium had not been reached. The question of introduction of material will be covered below.

The second case is that of an open system, first discussed from the point of view of the phase rule by Korzhinsky (1936). In this situation, the system of interest can be thought of as being associated with a reservoir of very large size in comparison to the dimensions of the system. Certain "mobile" components are able to move between the reservoir and the system. The chemical potentials of these components can be assumed fixed by the reservoir and are, then, no longer variable unknowns in the phase rule. The chemical potentials of those components of the system which are not mobile are still variable unknowns. The phase rule in this case gives: minerals equal immobile components. This appears to be a valid rule, as shown in known cases of metasomatism such as wall-rock alteration and corrosion of metals.

Returning to the schist-quartz vein example, if the silica in the vein is considered as introduced, then SiO_2 is a mobile component. Thus, the total of immobile components is four, assuming that water is probably also mobile in this case. Thus phases outnumber immobile components. Again assuming that equilibrium has been reached, this indicates that silica was not introduced and that all the minerals formed in a closed system as shown above, with the quartz vein representing a fluid silica phase formed from the original sediment during the intense metamorphism. This type of approach can be made to any problem of mineralogical association. However there are definite limitations in using these phase rules owing to the difficulty of adequately defining a given system with respect to chemistry and relations between phases. (See Turner and Verhoogen; 1951, p. 423.)

A possible application of the open system phase rule is in studying the suggested process of granitization. For instance, granite bodies in association with quartzite are often postulated as originating by granitization. If a quartzite body is considered as the system and a reservoir of granitic material is postulated "at depth", then the supposed process would involve introduction of material, transforming part of the quartzite into granite. The open system phase rules gives: minerals equal immobile components. A possible granite might have as few as three minerals, while the number of immobile components can be as low as one.

This means that the greatest possible number of minerals which could form at equilibrium is one. If equilibrium has been attained it was not in the manner given here, since minerals outnumber immobile components. Assuming the open system phase rule to be valid, this reasoning throws doubt on the supposed processes of granitization, especially in the case of bodies of limited chemical complexity such as quartzite, or in cases where movement of almost all material as granitic and basic "fronts" is postulated, since the number of minerals resulting would have to be equal to the small number of immobile components.

The open system phase rule can also be applied to the thin monominerallic and biminerallic bands often found at the contacts of ultramafic bodies. These bands are generally attributed to material introduced by hydrothermal solutions. This could be considered as a case of the most mobile components from a reservoir coming in furthest from the outside toward the center of a system, defined as an ultramafic rock, and forming monomineralic zones by the requirements of the phase rule. Thus the different zones would be a measure of the relative mobility of the different chemical species. The one immobile (i.e., not introduced and not moving) component of the system would be the MgO of the dunite, peridotite, or serpentinized equivalent thereof. FeO, if present in significant amount, could also be considered immobile, producing a tendency to bimineralic zoning.

A review of the literature (see Appendix A) indicates that banding of this type tends to form the following sequence at contact

areas: ultramafic rock (including serpentine), talc, tremolite-actinolite, chlorite, biotite, country rock. In general this appears to be true no matter what the country rock may be. These bands can be considered to represent the introduction of SiO_2, CaO, Al_2O_3, and K_2O, in order of decreasing mobility, as shown by the following equations:

1) $Mg_3Si_2O_5(OH)_4 + 2SiO_2 = Mg_3Si_4O_{10}(OH)_2 + H_2O$

Serpentine — Talc

2) $5Mg_3Si_4O_{10}(OH)_2 + 6CaO + 4SiO_2 = 3Ca_2Mg_5Si_8O_{22}(OH)_2 + 2H_2O$

Talc — Tremolite

3) $Ca_2Mg_5Si_8O_{22}(OH)_2 + Al_2O_3 + 3H_2O = Mg_5Al_2Si_3O_{10}(OH)_8 + 2CaO + 5SiO_2$

Tremolite — Chlorite

4) $6Mg_5Al_2Si_3O_{10}(OH)_8 + 5K_2O + 12SiO_2 =$

Chlorite

$$10KMg_3AlSi_3O_{10}(OH)_2 + Al_2O_3 + 14H_2O$$

Phlogopite

For olivine instead of serpentine:

1a) $3Mg_2SiO_4 + 5SiO_2 + 2H_2O \rightleftharpoons 2Mg_3Si_4O_{10}(OH)_2$

Olivine — Talc

Often anthophyllite is found instead of, or with, tremolite-actinolite, indicating not enough CaO as compared to MgO:

2a) $Mg_3Si_4O_{10}(OH)_2 + 4SiO_2 + 4MgO \rightleftharpoons Mg_7Si_8O_{22}(OH)_2$

Talc — Anthophyllite

3a) $5Mg_7Si_8O_{22}(OH)_2 + 7Al_2O_3 + 23H_2O \rightleftharpoons 7Mg_5Al_2Si_3O_{10}(OH)_8 + 19SiO_2$

Anthophyllite — Chlorite

Water is undoubtedly mobile and probably is the main component of the altering solutions. Iron oxide has been omitted from the above equations for simplicity and because it is usually present only in small quantities in the contact minerals. Thus the biotite band should perhaps be referred to as a phlogopite band. When carbonate is found with these rocks it is usually associated with talc, and also replaces serpentine, probably indicating that Co_2 has a strong mobility. The most common and extensive alteration of ultramafic rocks found, often called steatitization, results in formation of zones or large masses of talc-carbonate rock, indicating that SiO_2 and carbon dioxide usually represent the only extensively introduced material other than water.

In most cases the above ideal sequence is not found and many variations occur, usually with one or more of the zones missing. Phillips and Hess (1936) have suggested that the banding is a reflection of control by falling temperature and postulate a biotite-actinolite high-temperature form, and a chlorite-talc low-temperature form which is often impressed on the earlier high-temperature effects. As outlined above, the process is viewed as one of relatively simultaneous formation of zones, with the varying results representing the different combinations of such variables as amount of introduced material, length of time during which alteration occurred, permeability of ultramafic matrix, temperature, etc. Partial replacement of one zone by another would thus represent shifting equilibrium as material continued to be introduced.

The end result to which the process would tend would be a single monomineralic band (or a single bimineralic band if FeO is a major component of the ultramafic) and this together with non-availability of certain elements may explain the many cases where only one or two of the bands occur.

CONDITIONS FOR THERMODYNAMIC EQUILIBRIUM

In the next chapter chemical analyses of minerals separated from a serpentine contact area will be presented and their formation will be discussed in the light of these data. If this formation is a result of local interaction between two chemically different rock types, as some geologists have suggested, rather than of introduction of new material by hydrothermal solutions, as outlined above, then necessary and sufficient conditions of the equilibrium state to which this process would tend can be outlined. Although the possible attainment of these conditions cannot be tested with respect to temperature and pressure, it can be partially tested with respect to composition. This has been the purpose of the chemical analyses. This section is included to indicate briefly the theoretical basis for the experimental work.

Let us first consider a system made up of homogeneous pelitic schist. The system will be assumed to have been closed during metamorphism and to consist of the material now found in it. We will further define the system by assuming that the total volume has been constant and that it has been adiabatically insulated

and, therefore, of constant total entropy. Thermodynamic considerations show that the condition for a system of constant total entropy, volume, and composition to be in equilibrium is that the internal energy be at a minimum. Considering the internal energy as a function of several variables, this condition can be expressed mathematically in terms of these variables as necessary and sufficient conditions of equilibrium. The necessary conditions are based on the first derivative requirements of a minimum for a function of several variables, and the sufficient conditions on the second derivative requirements. Using the method of Lagrange multipliers, it can be shown that the necessary conditions for the internal energy to be a minimum are:

1) that the temperature of all phases be equal: $T' = T^2 = \ldots T^\alpha = C$;

2) that the pressure on all phases be equal: $P' = P^2 = \ldots P^\alpha = C'$;

3) that the chemical potential μ_i of any component i be the same in all phases in which it is present: $\mu_i' = \mu_i^2 = \ldots \mu_i^\alpha = C''$.

Condition three was used earlier in developing two phase rules, both of which are based on the assumption of attainment of equilibrium. Here we are using different variables and are dealing with the internal energy instead of the Gibbs free energy, so that the chemical potential μ_i is expressed as: $\mu_i = \left(\frac{\partial U}{\partial n_i}\right)_{S,V,n_j,n_k,\text{etc.}}$

It can be further shown that sufficient conditions for equilibrium are:

1) that the temperature increase with an increase in entropy (thermal stability): $\left(\frac{\partial S}{\partial T}\right)_V > 0$;

2) that the pressure decrease with an increase in volume (mechanical stability): $\left(\frac{\partial V}{\partial P}\right)_S < 0$;

3) that the chemical potential μ_i of any component i in a phase increase with an increase in the amount of the component in the phase: $\left(\frac{\partial \mu_i^\alpha}{\partial n_i^\alpha}\right)_{SV} > 0$.

Thus, assuming a given geologic process has occurred in a system which could be defined as above, and assuming this process reached thermodynamic equilibrium, the resulting rock mass would be characterized by the specified necessary and sufficient conditions of equilibrium. Obviously none of the pressure and temperature requirements can be checked with respect to rock formation. However the necessary condition for composition can be checked if ideal conditions are assumed. The other conditions have been outlined to give a complete picture of the equilibrium requirements. By placing different restrictions on a system various other equivalent conditions can be developed with respect to variables other than temperature and pressure, but the composition requirements remain the same. The special case of the existence of a fluid pressure as well as a solid pressure does not alter any of the general requirements.

To outline a means of checking attainment of equilibrium in a rock such as the schist described above, let us consider the biotite grains in the schist. The various minerals in the schist represent different phases, while the biotite grains are different examples of the same phase, namely: $K(Mg,Fe)_3AlSi_3O_{10}(OH)_2$.

In particular we are interested in the Mg/Fe ratio of the different grains. If we assume that the magnesium and iron ions in a biotite grain represent an ideal mixture, i.e., that either ion can substitute equally well in the structure, then the chemical potentials of magnesium and iron in the grain can be expressed as: $\mu^{\alpha}_{Fe} = \mu^{\circ}_{Fe} + RT \log \bar{n}^{\alpha}_{Fe}$

and: $\mu^{\alpha}_{Mg} = \mu^{\circ}_{Mg} + RT \log \bar{n}^{\alpha}_{Mg}$

where μ°_{Fe} refers to the chemical potential of iron in the pure state, μ°_{Mg} to the chemical potential of magnesium in the pure state, and $\bar{n}^{\alpha}_{Fe}$ refers to the mole fraction of iron based on the total number of moles found in six-fold co-ordination, $\bar{n}^{\alpha}_{Mg}$ to the mole fraction of magnesium on the same basis.

The assumption of ideal behavior is necessary to avoid use of activity coefficients, which give a quantitative measure of the discrepancies between actual behavior of a mixture and so-called "ideal" behavior. Ideal behavior is defined by certain thermodynamic relationships and represents the simplest possible behavior. We have very little information on the degree of ideality of mineral solid solutions, and no numerical values for the pertinent activity coefficients. The assumption of an ideal mixture for the magnesium and iron ions in biotites is thus an approximation. The two ionic radii do not differ greatly in size, a necessary condition for ideal behavior.

Assuming equilibrium has been attained in the schist, the necessary conditions of this state require that for two biotite

grains: $\mu_{Fe}^{\alpha} = \mu_{Fe}^{\beta}$ and $\mu_{Mg}^{\alpha} = \mu_{Mg}^{\beta}$.

Therefore $\bar{n}_{Fe}^{\alpha} = \bar{n}_{Fe}^{\beta}$ and $\bar{n}_{Mg}^{\alpha} = \bar{n}_{Mg}^{\beta}$.

Thus, if we could analyze two separate biotite grains which have been formed in equilibrium with each other, the mole fraction of iron in each grain should be the same, as should be the mole fraction of magnesium. Naturally, in actual chemical analysis a number of grains from one sample must be compared with a number of grains from another sample.

However, other elements, such as aluminum, may substitute for iron and magnesium, and these other elements may not behave ideally with respect to each other. In this case, even though

$\mu_{Fe}^{\alpha} = \mu_{Fe}^{\beta}$ and $\mu_{Mg}^{\alpha} = \mu_{Mg}^{\beta}$, $\bar{n}_{Fe}^{\alpha} \neq \bar{n}_{Fe}^{\beta}$ and $\bar{n}_{Mg}^{\alpha} \neq \bar{n}_{Mg}^{\beta}$.

For this situation the mole fractions of iron and magnesium should be based on the total number of moles of iron and magnesium only. A convenient method of comparison is then mole fraction ratios, with equilibrium requiring: $\bar{n}_{Mg}^{\alpha} \Big/ \bar{n}_{Fe}^{\alpha} = \bar{n}_{Mg}^{\beta} \Big/ \bar{n}_{Fe}^{\beta}$.

Thus, attainment of equilibrium can still be checked by measuring the Mg/Fe ratios in biotite grains. In actual calculations a basis of 12(O,OH) atoms can be used to compute the number of Mg and Fe atoms represented by a given analysis, and the ratio of these numbers can be used as the equilibrium check.

This reasoning might be applied to biotite grains in a monomineralic band at a contact. However the assumption must be made

that the banding formed in a closed system, and the above volume and entropy conditions would also have to hold. In this case the system would be defined by the contact and would include all material on either side of the contact which had been altered by interaction across the contact. Earlier, a possible explanation for the banding was presented which treated the contact as a permeable boundary of an open system, the ultramafic rock, with outside material introduced into the system. Here, it would be assumed that no addition from a source away from the contact has taken place, and the banding is viewed strictly as a contact reaction between two different, homogeneous rock types. Also, it is probable that equilibrium would occur only at the contacts between such bands, with a gradation in chemical composition of the minerals occurring perpendicular to the contact in each band. A constant Mg/Fe ratio in co-existing biotite-actinolite samples at different sites along their mutual contact would indicate attainment of equilibrium at the contact. In order to test the variation of mineral composition in such contact bands, a number of analyses of biotite and actinolite samples from a contact area have been carried out. These are presented and discussed in the following chapter. Readers interested in a further discussion of equilibrium in metasomatic processes are referred to a paper by Thompson (1959).

CHAPTER V. ANALYSIS OF CONTACT MINERALS

INTRODUCTION

Twelve biotite and seven actinolite samples from adjacent bands of these minerals at the contact between quartz diorite and serpentine-marble have been analyzed in duplicate by rapid analysis methods. The location of these samples, which are all from the southern quarry, is shown in Figure 9. The banding is pictured in Figure 10. Unfortunately, owing to a lack of adequate fresh samples in the field, the sampling is of a somewhat random nature. Ideally, several series of samples in each band perpendicular to the contact should be analyzed, since it is likely that a variation in chemical composition of the two minerals occurs perpendicular to the contact. Another departure from an ideal study of these bands is the variation in the two wall rocks found here, with some pelitic schist replacing the quartz diorite at one area and with the serpentine-marble varying from pure serpentine to marble with essentially no serpentine. However the analyzed samples do give a quantitative idea of the amount of compositional variation which may be found in the two minerals in these bands. Further, among the samples analyzed there are five pairs of co-existing biotite-actinolite taken at different distances from the contact between the two bands. The analyses of these pairs gives an indication of the direction of movement of the various elements and of the relation of the composition of the minerals to distance from the contact. Finally, this work gives an example of the precision and accuracy of rapid analysis

procedures as applied to minerals, since all samples were analyzed in duplicate and the standards, G-1, W-1, and Haplogranite, were run at the same time and under the same conditions.

Relatively little work has been published on the variation in chemical composition of tremolite-actinolite, while biotite has received as much attention as any mineral. Hall (1941) discusses the relation between color and composition in biotites, and presents fifty-six analyses of biotites for MgO, FeO, TiO_2, and Fe_2O_3, mostly taken from the literature up to 1941. Heinrich (1946) gives seventeen analyses of biotites representing the maximum content of the various constituents which may occur in the mineral. A proposed relation between chemical composition and paragensis in biotites of igneous rocks is outlined by Nockolds (1947). He gives a total of seventy-seven biotite analyses. A broad study of all the micas can be found in Henrich et al. (1953).

PREPARATION OF SAMPLES

From each field sample a rough cube approximately 1" on a side was cut. The cube was then broken up by mortar and pestle. That part of the sample which passed through a 60 mesh screen, but not through a 140 mesh screen, was kept. Next, the sample was purified to 99% or better by as many as twenty passes through a Frantz Isodynamic Magnetic Separator. Purity and identity of impurities were checked by sample immersion in refractive index liquids. Most of the samples were found to be over 90% pure even before use of the separator, because of selective field sampling. Minerals other than biotite and actinolite found in one or more

samples were apatite, talc, magnetite, muscovite, tourmaline, and andesine. Most of these were easily separated, as was biotite from actinolite, once the correct combination of separator angle and field strength was found.

All of the actinolite samples had the same beta index ($1.630 \pm .003$) as determined in white light. Similarly, all the biotite samples with the exception of #172 had a beta=gamma index of $1.597 \pm .003$. For #172, geta=gamma = $1.608 \pm .003$. Part of the biotite grains in most of the samples exhibited a few very fine-grained pleochroic haloes. The actinolite was colorless to very light green and slightly pleochroic from light green to light blue-green. The biotite was found to be very light grey-green and non-pleochroic, again with the exception of #172, which was light to dark brown and non-pleochroic.

PROCEDURES

The analytical procedures which were used are based on the work of Shapiro and Brannock (1956). A somewhat different scheme of analysis has been published by Riley (1958a). This recently developed type of chemical analysis of silicate rocks and minerals is referred to as rapid analysis, as distinguished from conventional chemical methods. Perhaps a better name would be instrumental analysis, because this is the feature which characterizes most of the procedures. Modifications of these procedures for samples with unusual compositions may result in a "rapid" analysis consuming more time than a conventional analysis. For example, some of the procedures have to be altered or cannot be

used if a high concentration of a particular element occurs, as is often the case in analyzing minerals. However the relative cheapness, small amount of sample needed, and rapidity of measurement will result in the use of rapid analysis for studying geologic problems in the future when otherwise time requirements and cost would prohibit chemical work. The first published work of this kind known to the author is that of Pitcher and Sinha (1958). Other examples include the present work, and two other Ph.D. theses recently carried out at M.I.T. (Kranck - 1959, Phinney - 1959).

Fortunately, the methods of Shapiro and Brannock (1956) could be used without alteration for the work presented here. SiO_2, Al_2O_3, Fe_2O_3, TiO_2, P_2O_5, and MnO were determined by measuring with a spectrophotometer the transmission of light through sample solutions. Ferrous iron was found by the conventional titrimetric method. The automatic photometric titration technique using a spectrophotometer and recorder, described by Shapiro and Brannock (1956), was used for CaO and MgO. However they have recently improved the chemical part of this procedure by the use of an organic complexing agent, and kindly provided our laboratory with the necessary details so that this new procedure could be used. K_2O and Na_2O were measured on a Perkin-Elmer Model 146 flame photometer using lithium as an internal standard. .85 grams of sample were used in a single analysis for the above elements, with .40 grams of this amount used for the ferrous iron determination. In order to drive off all of the water from minerals such

as micas and amphiboles it is necessary to have special equipment, since temperatures on the order of 1200°C must be reached. Thus the modified Penfield method used by Shapiro and Brannock (1956) could not be used for this work. This is, of course, unfortunate because the standard chemical check of addition to 100% is eliminated. Numerous analyses of garnets carried out in the M.I.T. laboratory did give a good 100% check (Phinney, personal communication). A calculation of mineral formulas from the analyses given here is still possible, and the results of such calculations may be found in Appendix C. The water content of G-1 and W-1 was determined using the modified Penfield method. (For a discussion of the problem of water analysis see Groves - 1951, pp. 95-104. Riley - 1958b, has recently given a quantitative comparison of various methods of water determination as applied to minerals.)

PRECISION AND ACCURACY

A discussion of advantages and results obtainable in rapid analysis of silicate rocks is given by Shapiro and Brannock (1956). Mercey (1956) presents a detailed study of the accuracy and precision of the rapid methods as applied to rock analysis. He concludes that the accuracy and precision of these methods compares favorably with results obtainable by conventional methods. The author concurs with this conclusion, provided duplicate samples are analyzed and each sample is run in duplicate. The latter qualification adds relatively little to the total time required for the rapid techniques, and is a necessity since aberrant results

do occur frequently enough to be significant. This concurrence applies only to analyses of samples with element concentration ranges of common igneous rocks. A further qualification must be made for the application of these methods to mineral analysis. In this case unusually high or low concentrations of various elements occur, and results comparable to those of conventional methods may not be obtainable. Mercey also points out the lack of a precise and accurate method for water determination. A thorough interlaboratory study of precision and accuracy in chemical analysis of silicate rocks has recently been made (Fairbairn et. al. - 1951). Further discussion of this topic is given by Fairbairn and Schairer (1952) and by Fairbairn (1953).

The results of the analyses of the three standards and of the biotite and actinolite samples, and the calculation of these latter results to an atomic basis, are given in Appendices B and C. Also in Appendix B are published analyses of seven contact biotite samples and one contact actinolite sample. These analyses are the only ones of contact samples of these two minerals which could be found in the literature. The biotite and actinolite samples analyzed by Phillips and Hess (1936) were taken at the contact between serpentine and schist at Chester, Vermont, and have also been calculated to an atomic basis to provide a comparison with the work reported here. To provide another comparison, two commercial laboratories were sent two grams of biotite #210 for analysis. One laboratory (Ledoux and Co.) requested and received five more grams of sample. The results reported by these laboratories are also included in Appendix B.

DISCUSSION OF RESULTS

The significance of the mineral analyses reported here can be best discussed from two viewpoints. Firstly, some individual and general features should be mentioned. Secondly, and more importantly, the composition of the co-existing biotite-actinolite pairs can be studied to provide an idea of the chemical changes which have occurred at the contact. Problems of interest here are direction and degree of movement of the various elements; amount of compositional variation to be found, including the variation in the Mg/Fe ratios of the two minerals; and degree of approach to chemical equilibrium, including adjustment both across and along the contact.

Before discussing the significance of the analyses, some further comments on their accuracy should be made. The calculations to an atomic basis given in Appendix C were made by assuming the difference between the total of the chemical work and 100% to be all water. Only if no other elements are present in significant amount is this assumption valid. The main possibility here is fluorine. The analysis of biotite #210 by Ledoux and Co. indicates a low fluorine content for these minerals. The high fluorine content reported by Booth, Garrett, and Blair is obviously in error, as indicated by their extremely low silica value. The totals of the various analyses fall into a limited range for each mineral, and agree with totals to be expected for all major elements other than water. The total for actinolite #161 is low, and may indicate the presence of another major constituent.

The calculated atom values given here show good agreement between the chemical analyses and the requirements of the crystal chemistry of micas and amphiboles. For example, the sum of the cations of an amphibole should fall between fifteen and sixteen, and this value varies from 14.96 to 15.51 for five of the analyzed actinolites. The total for #175 is 14.64 and for #161, which has the low summation, 13.88. The analyses of the three standards provided another check on the mineral analyses, with results for the standards being similar to those obtained by conventional methods. The analyses of biotite #210 by the two commercial laboratories show wide disagreement in the results for several elements and neither laboratory shows close agreement with the author's values. This points out the importance of evaluating a mineral analysis by a crystal chemistry check.

Not unexpectedly, a rather large variation occurs in the composition of the two minerals. The major element variations in the biotites are SiO_2: 37.93 - 42.05%; Al_2O_3: 14.26 - 19.22%; FeO: 2.90 - 7.96%; Fe_2O_3: 1.13 - 3.04%; MgO: 17.98 - 24.42%; and K_2O: 8.96 - 10.31%. In the actinolites the variations are SiO_2: 51.33 - 58.34%; Al_2O_3: .02 - 7.70%; FeO: 2.28 - 3.90%; Fe_2O_3: .56 - 1.74%; MgO: 19.49 - 24.90%; and CaO: 10.04 - 13.34%. These variations, most of which are too great to be attributed to experimental error, may indicate non-attainment of chemical equilibrium, the effect of varying wall rock and source of material, or both. They do indicate that only one or two analyzed specimens are an inadequate sample of the composition of such contact minerals.

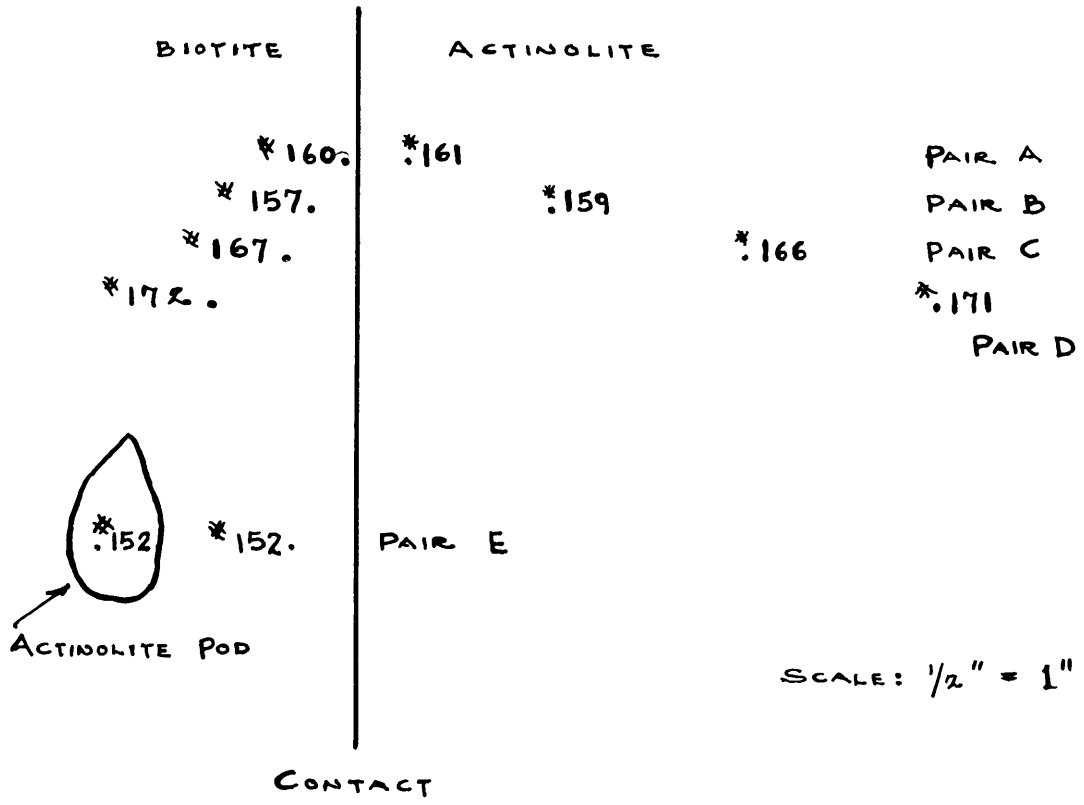
Another indication of the chemical variation present in these monomineralic bands is given by comparing those samples which were taken close together. Biotites #157 and #158, taken about one foot apart, show a 1% variation in silica and a 2% variation in alumina. This may arise from the presence of a small feldspar pod in the mica band one inch from #158, while #157 is a representative sample of the band. Biotite samples #206 - #208 and #209 - #210 were taken approximately six feet apart and show what appear to be significant differences in their content of certain elements.

Biotite samples #167 and #172 were taken where pelitic schist replaces quartz diorite as wall rock of the bands. The only difference in chemical composition of these samples is their high content of TiO_2 as compared to that of the other biotites. In the case of #172, which has 1.14% TiO_2, the color and index of the mineral differ from the other biotites, apparently because of the titania content. No change in titania content is shown by the co-existing actinolite samples of these two biotites.

High contents of CaO and P_2O_5 in biotite samples #152 and #153 would seem to indicate appreciable apatite impurity. However the samples were carefully checked with refractive index liquids and no apatite was found.

A schematic representation of the relation of the analyzed biotite-actinolite pairs to the contact between the two monomineralic bands is shown in Figure 17. Figure 18 shows the variation in composition with respect to the contact between the two

Representation of Location of Analyzed Biotite - Actinolite Pairs with Respect to Contact Between the Two Monomineralic Bands



PAIR A — APPROXIMATE DISTANCE APART PERPENDICULAR TO CONTACT: 5/8"

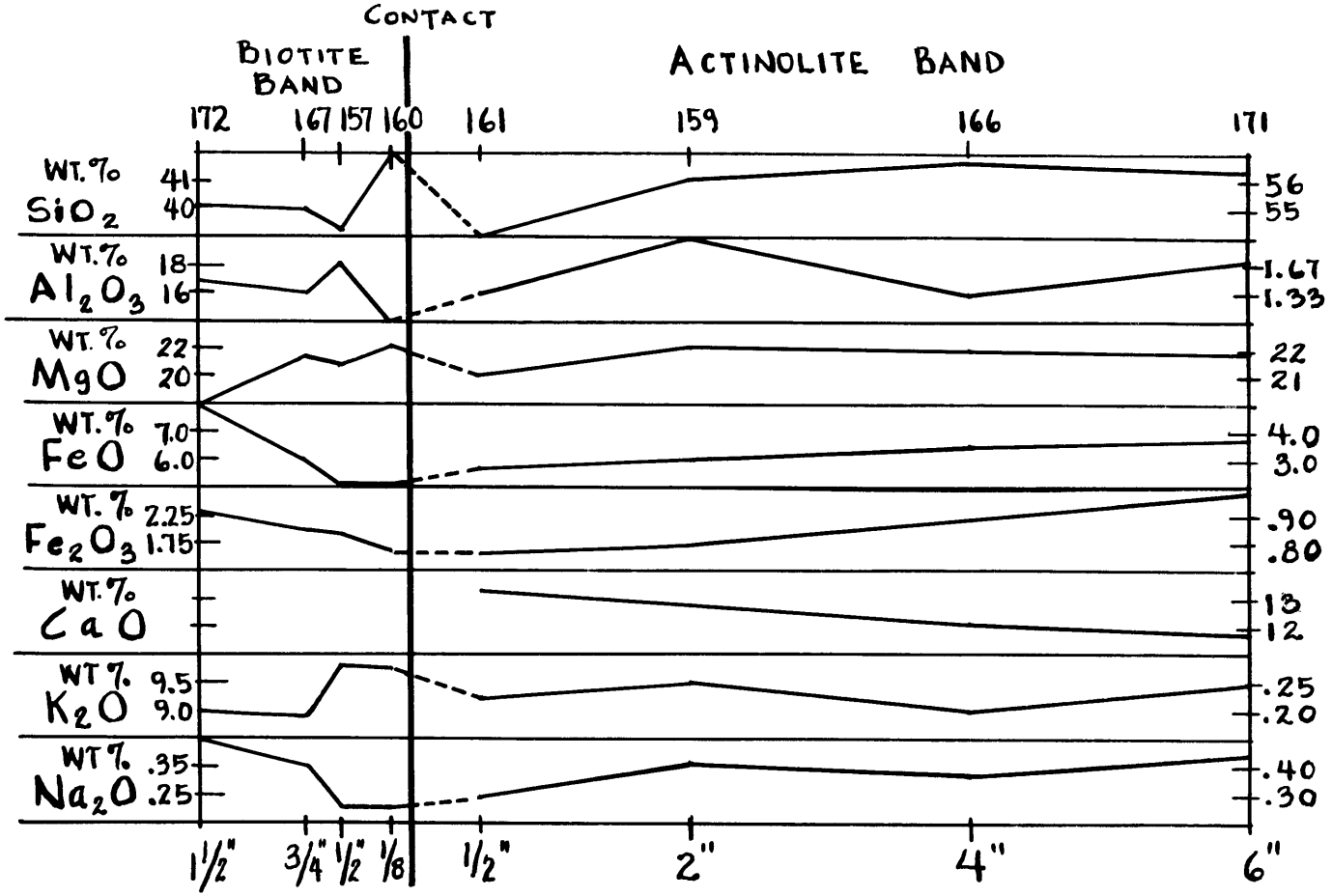
PAIR B — " " 2 1/2"

PAIR C — " " 4 3/4"

PAIR D — " " 7 1/2"

PAIR E — " " 2"

FIGURE 17.

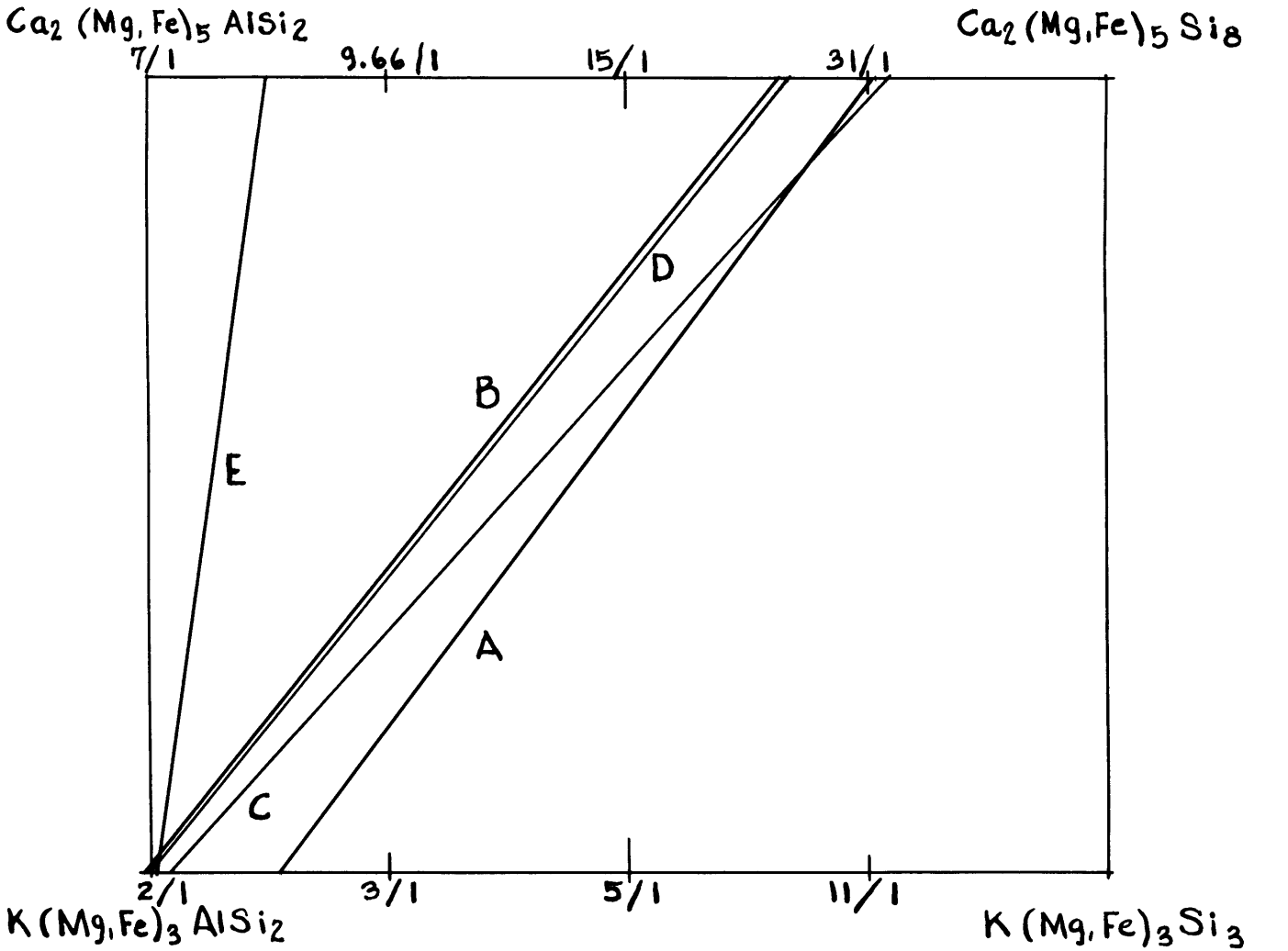


MINERAL COMPOSITIONAL VARIATION WITH RESPECT TO BIOTITE–ACTINOLITE CONTACT

FIGURE 18.

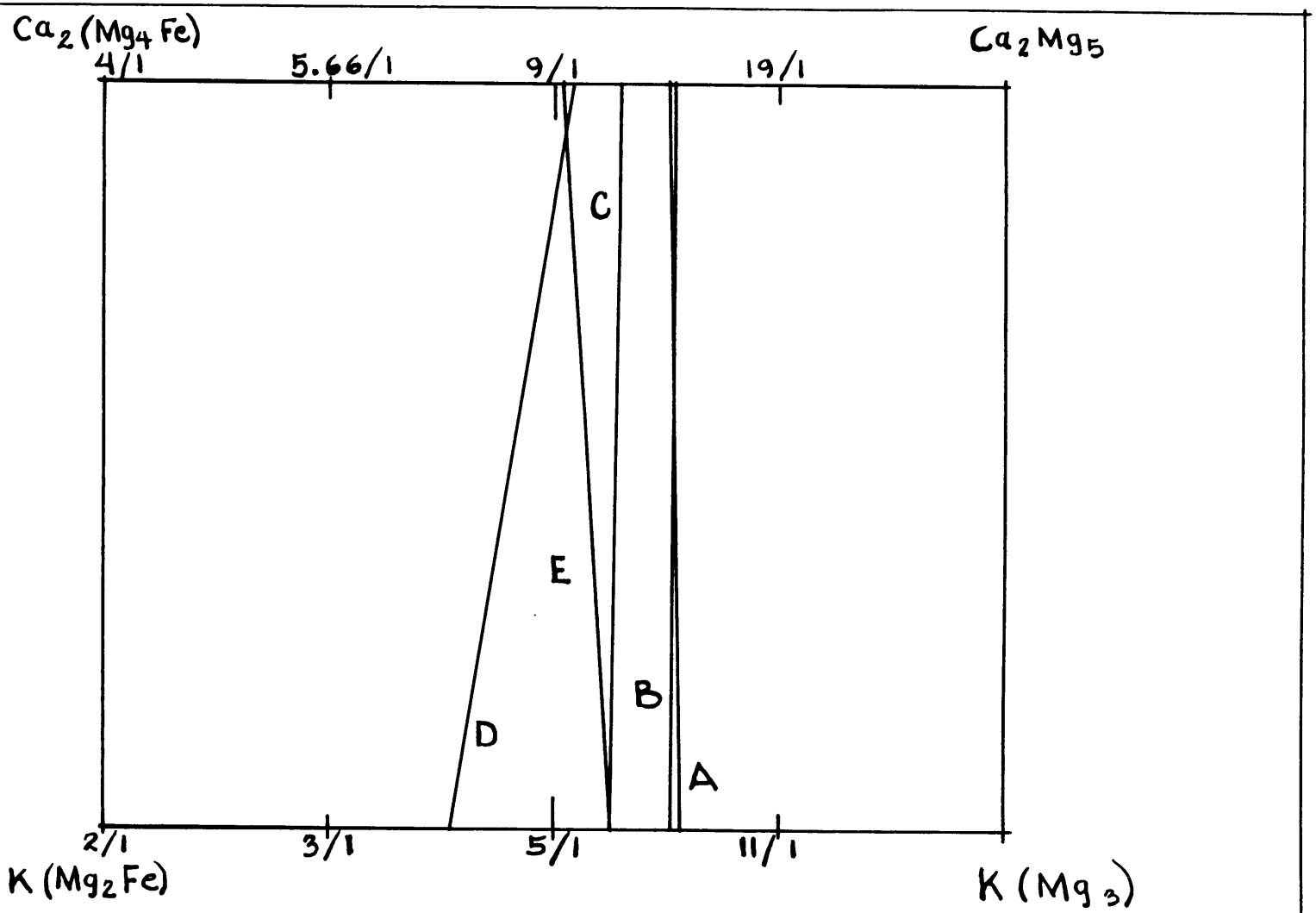
bands. As pointed out earlier, the four mineral pairs of Figure 12 do not actually come from one line across the two bands. Each pair is from a different point along the contact, with one pair from another contact, as has been shown in Figure 9. It is likely that Figure 12 represents the kind of variation which exists perpendicular to the contact. This is seen as culminations and depressions in the element content of the two minerals in the vicinity of the contact. A similar result was found by Dennen (1951) at the contact between certain intrusives and their wall rock. He states: "all major rock constituents studied (Al, Fe, Mg, Mn, Ca, Na, and K) are mobile when moderate temperatures and concentration gradients are present." Further, "highly irregular variations are found when the intrusive and wall rock were compositionally different and steep thermal gradients existed. These latter variations may show geochemical culminations and depressions that are wave-like in form and die out in both directions from the contact zone." This work indicates that the variations found by Dennen in contact rocks exist also in contact minerals.

Figures 19 and 20 show the relationship of the Si-Al and Mg-Fe atomic ratios of the co-existing biotite-actinolite pairs. It is interesting to note that Pair E, which was taken at the contact of an actinolite pod in the biotite band, and therefore represents a definite disequilibrium situation, has a much different slope in both diagrams as compared to the other pairs, which were taken across the main contact between the two bands.



Si-Al Atomic Ratios for Co-Existing Biotite – Actinolite Samples

Figure 19.



Mg-Fe ATOMIC RATIOS FOR CO-EXISTING BIOTITE-ACTINOLITE SAMPLES
FIGURE 20.

Assuming that Mg and Fe have been introduced from the serpentine-marble, and Si and Al from the quartz diorite-schist, the change in slope of the pairs away from the main contact indicates silica to be more mobile than aluminum and iron to be more mobile than magnesium. The variation in the Mg/Fe ratios in the two minerals indicates a degree of chemical disequilibrium in the bands, since it has been shown in Chapter IV that attainment of equilibrium requires that the Mg/Fe ratios in different samples of the same mineral be similar. (See Fig. 21).

Calculated Mg/Fe Atomic Ratios In Biotite-Actinolite Pairs

	Pair A	Pair B	Pair C	Pair D	Pair E	
	#160	#157	#167	#172	#152	
Mg	2.338	2.206	2.273	1.917	2.121	Biotite
Mg/Fe	7.96	7.43	6.40	4.03	6.39	
Fe	.294	.297	.355	.475	.332	
Biotite Mg/Fe / Actinolite Mg/Fe	.622	.567	.569	.393	.699	
Mg	4.075	4.506	4.495	4.482	4.180	Actinolite
Mg/Fe	12.81	13.10	11.24	10.25	9.14	
Fe	.318	.344	.399	.437	.457	
	#161	#159	#166	#171	#152	

Figure 21

Additional Note

Since completion of this manuscript the author has come across a recent publication by Kretz (1959). Kretz presents sixty-two spectrochemical analyses of co-existing biotite, garnet, and hornblende from Grenville gneisses of Quebec and discusses the results in terms of phase equilibrium. In texting the attainment of thermodynamic equilibrium he emphasizes use of the distribution coefficient for a given element between different minerals. It is concluded that a close approach to equilibrium was reached in these rocks.

Kretz, R. (1959), Chemical Study of Garnet, Biotite, and Hornblende from Gneisses of Southwestern Quebec with Emphasis on Distribution of Elements in Coexisting Minerals, Jour. Geol., v. 67, p. 371-402.

APPENDIX A

Brief Survey of Similar Areas

Almost all serpentine occurrences described in the literature have associated amphibolite rocks and, sometimes, associated limestone or dolomite beds. Usually one or more carbonate minerals occurs with the serpentine rocks. Also many of the serpentine bodies are small in size, parallel the regional structure, and have a small content of chromium and nickel. Many exhibit monomineralic banding at contacts with country rock. Those which differ from the western Massachusetts bodies have a significant amount of olivine, or of olivine and pyroxene, and provide abundant and definite evidence of their derivation by alteration of these minerals, thus indicating an igneous origin. The areas reviewed in the following pages have been selected for their similarity to the serpentines of western Massachusetts, or for the detailed study of contact banding given by the author. Those interested in further descriptive material are referred to the extensive literature on the geology of the Alps. Three papers dealing with Vermont ultramafic bodies, which represent the northern extension of the western Massachusetts belt, will be summarized, followed by a description of some areas in other parts of the world.

Chidester et. al. (1951) classify Vermont ultrabasic rocks in two types: 1) verde antique -- completely serpentized and more or less extensively steatitized; 2) partly serpentized dunite or periodotite and minor pyroxenite, commonly containing more or less chrysotile asbestos. These bodies range in width from a few

feet to a mile, and in length from less than one hundred feet to 13.5 miles. Most of them are emplaced in greenstone or amphibolite or in schist and phyllite in the immediate vicinity of greenstones or amphibolites. A typical body of type one has a core of serpentine, then several feet to a few tens of feet of grit (talc and carbonate), then an outer zone of a few inches to a few feet of talc with practically no carbonate. This outer zone may have some chlorite. Blackwall of chlorite or biotite, representing altered country rock, occurs at the outer border of the talc zone. The country rocks are phyllites, schists, and gneisses, with intercalated schistose greenstones and amphibolites. The latter represent altered volcanic tuffs and flows and intrusive rocks consisting chiefly of sill-like dikes. Mafic dikes, formed after regional folding and metamorphism, intrude the ultrabasic rock and country rock at several localities. There are no known granitic intrusive rocks, other than small felsic dikes at a few localities, associated with the ultrabasic bodies. The probable age range of the intruded formations is Cambrian to Middle Ordovician.

Gillson (1927) describes and discusses a number of Vermont talc deposits. Most of the deposits occur in quartz-sericite schist, with talc intimately associated with ferrous dolomite. Some are associated with serpentine, but no workable talc occurs wholly in serpentine. All of the deposits are bounded by a few inches of pure chlorite (blackwall), which is believed to represent a stage in the alteration of the country rock to talc.

Associated with the blackwall are tourmaline, biotite, actinolite, apatite, magnetite, titanite, and pyrrhotite. At one deposit studies indicated the biotite and chlorite of the country rock to be optically different from the biotite and chlorite of the talc zone. Gillson (p. 274) believes the deposits "are a kind of contact metamorphic deposit, formed by hot alkaline solutions, low in silica, and toward the last rich in magnesia and CO_2." These solutions formed talc by replacing both schist and serpentine. The sequence of mineral formation given is: tourmaline, quartz, biotite, apatite, amphibole, chlorite, titanite, dolomite, talc, magnetite, and pyrrhotite. He points out that talc occurs with similar minerals in both metamorphosed limestone and in altered ultrabasics and gives a number of examples of the association of talc with granitic rocks, although no evidence for a granitic source for the talc-forming solutions was found in Vermont.

Phillips and Hess (1936) discuss contact effects between serpentine and siliceous country rocks as shown in the northern Appalachians. Two main types are distinguished: 1) the low-temperature type is marked by formation of a band of chlorite at the edge of the country rock schist and of talc at the edge of the serpentinite; actinolite is found in both the chlorite and talc zones, and talc may replace it; the original contact is thought to have been between the talc and chlorite zones: 2) the high-temperature type has a band of biotite at the edge of the country rock and of actinolite at the edge of the serpentinite;

the original contact is thought to have been between the biotite and actinolite. These monomineralic zones are measured in inches, and grade into the adjoining rock within a few inches. Usually low-temperature alteration occurs after the high-temperature effects. This is shown by replacement of biotite by chlorite and of actinolite by talc. The authors feel these effects are due to hydrothermal solutions which allowed migration of material across the contacts with introduction of silica, alkalies, carbon dioxide, flourine, boron, and sulfur. They point out that pegmatites and small felsic intrusions are generally abundant in those areas showing the higher temperature type of metamorphic differentiation and are probably the source of at least a large part of the hydro-thermal solutions.

Read (1934) deals with small round bodies of rock from the Shetland Islands which are made up of circular monomineralic zones. The largest mass is twenty feet in diameter. These rocks occur in gneisses of varying mineralogy which have been intruded and permeated by abundant acid injections. The pelitic gneisses have garnet, staurolite, kyanite, and biotite, with later chlorite and chloritoid, and with conspicuous feldspar blebs. Others are composed of biotite, muscovite, epidote, acid plagioclase, quartz, and iron oxide. Often found with the zoned bodies are hornblendic rocks of various types. The zoning tends towards the following ideal sequence from the center outwards: antigorite, talc, actinolite, chlorite, biotite, country rock. Both distorted

and undistorted types are found and both usually show fewer zones than the ideal case. The biotite contact with country rock is sometimes sharp, sometimes not. At transitional contacts oligoclase grains increase in amount until normal biotite-oligoclase gneiss is reached. The transitional portion is composed of biotite and muscovite with abundant epidote. This rock differs from the country rock only in the absence of plagioclase. Read suggests that the zoned masses originally consisted of peridotitic rock, with movement of material producing the zoning. The biotite zone is postulated to have originally belonged to the country rock and to mark the former contact between country rock and peridotite. He feels that these rocks cannot be explained on the Appalachain basis of successive formation of minerals with falling temperature and suggests that simultaneous formation has occurred here.

Macdonald (1941) discusses nodules of serpentine found in California enclosed in mica schist close to a body of intrusive quartz diorite. A large amount of amphibolite occurs in the area. The nodules have been altered to give the following sequence outward: serpentine, talc, talc and actinolite, actinolite, chlorite, and biotite. The serpentine core has criss-cross antigorite, relict rhombic pyroxene, metamorphic olivine, ultrabrown chlorite, ultrablue chlorite, and iron ore. The talc zone has talc, serpentine, both chlorites, and ore minerals. The talc-actinolite zone has felty talc, needles of actinolite, iron ore, and biotite. The biotite zone is ninety per cent biotite and ten per cent actinolite.

The country rock schist is mostly biotite with quartz, orthoclase, and oligoclase. The schist is distinctly richer in biotite than most of the schist of the region.

Pabst (1942) re-examined the nodules described by Macdonald (1941) and found several other minerals not mentioned by Macdonald. He also found that the changes of zoning varied greatly from nodule to nodule and in different parts of the same nodule. Some of the sequences which he found included: 1) talc, chlorite; 2) talc, anthophyllite, talc, tremolite, chlorite; and, 3) talc, anthophyllite, tremolite, vermiculite. Pabst (p. 570) concludes, "It is impossible to summarize these changes in a single scheme or graph." The actinolite and biotite of Macdonald were found by laboratory study to be mainly anthophyllite and vermiculite. The "mica schist" was found to have the composition of an igneous rock: 55% plagioclase, 25% hornblende, 18% biotite, 1% quartz, and 1% magnetite. He feels only two generalizations can be made about the zoning: 1) the innermost zone is always dominated by felted talc with only minor amounts of other minerals, which may include chlorite, magnetite, and monoclinic amphibole; 2) the outermost zone is always micaceous, but the character of the micaceous mineral varies.

Cairnes (1930) describes a serpentine belt in British Columbia which has gold in associated talcose shear zones. The main rock types are serpentine, greenstone (volcanic andesites), and argillaceous sediments, with the serpentine following what appears to

have been a zone of weakness in the associated rocks. The serpentine shows a persistent association with a particular greenish volcanic member of the rock series and the author, in an earlier general examination, concluded that the serpentine had probably developed through alteration of the greenstone. Later detailed field work indicated that the serpentine represents ultrabasics intruded along a zone of weakness coincident with a belt of greenish altered volcanics. This was followed and possibly accompanied by other intrusions less basic in character. The volcanic rocks vary in inverse ratio to the amount of serpentine. The sediment-volcanic contacts are transitional and it is often hard to distinguish the two. There are three main types of altered rock associated with the serpentine: 1) talc along shear zones; 2) carbonate-quartz-chrome mica; 3) talc and carbonate. The basic intrusives are altered to carbonate-talc-chlorite-serpentine rocks. At the contacts between greenstone and serpentine the serpentine is altered to talc and the greenstones to chlorite, with some further change to serpentine and talc.

DuRietz (1935) gives an extensive field, petrographic, and chemical study of ultramafic and altered ultramafic rocks of northern Sweden. The relatively unaltered rocks are mainly dunites and show no contact metamorphism. The ultramafics are almost invariably associated with amphibolite rocks, which occur everywhere in regional schists, generally as inconspicuous layers.

They are composed of green hornblende, plagioclase, zoisite or clinozoisite, rutile, sphene, ilmenite or magnetite, and quartz, and are often mica-bearing. Transitions appear between the schistose amphibolites and the mica-schists. Thin limestone beds occur locally. Where the amphibolites are scanty, the injection of granitic material is more prominent. The ultramafics are abundant along the main direction of folding axes and appear to have taken part in the folding. The serpentinized rocks have small bodies of pyrrhotite-chlorite rock scattered through them. Chrysotile occurs in the non-schistose serpentine bodies and antigorite in the schistose bodies.

The typical sequence at the schist-ultramafic contact is schist, chlorite, talc-actinolite, ultrabasic, with a similar sequence shown at amphibolite-ultrabasic contacts. The chlorite zone is sometimes missing. The following sequence is typical of the more complete type of transition: serpentinous dunite, serpentine-olivine-actinolite rock, talc-and actinolite-bearing serpentine, talc-actinolite rock, talc-chlorite rock, chlorite-actinolite rock, chlorite-anthophyllite-actinolite rock, chlorite rock, and amphibolite or vermiculite mica-schist. Secondary talc and carbonate are usual as transformations of earlier serpentine. The alteration products are best developed at ultramafic-amphibolite contacts. The best soapstone (talc-actinolite or talc-chlorite) deposits are localized where pegmatite is intruded. These pegmatites have a variable composition, with a low quartz content in contact areas of pegmatite and ultrabasic. In the pegmatites biotite may be formed at the expense of potash feldspar. The acid intrusions are held responsible for most of the

serpentinization and other alterations, while the ultramafics are believed to have been intruded "in a semi-crystalline condition." The amphibolitic rocks are concluded to be originally both intrusive and extrusive and associated with the same orogenic zones as the ultramafics.

Haapla (1936) describes serpentine rocks in Finland. These follow in strong conformity the enclosing rocks, although the contacts with country rock are seldom exposed. In one area serpentine gradually grades into dark amphibolite. Country rocks are mainly metamorphosed sediments, with intrusive granite in some areas. There is a complete lack of associated acid differentiates. Four exposed contacts show these sequences: 1) granite, a few inches of biotite, a few inches of chlorite, a few inches of talc with lesser carbonate, which rapidly grades into serpentine; 2) schist, biotite and chlorite, talc with tremolite next to the chlorite, serpentine with flakes of talc; 3) quartzite, diopside, tremolite, dolomite or talc, serpentine; 4) gneiss, black mica, chlorite, talc, ultramafic. Granitic veins in ultrabasics show this sequence: granite, brown mica, green chlorite, talc with carbonate and rare prisms of tremolite, transition to serpentine. At one contact of serpentine and granite there are patches of black mica and hornblende in the granite.

No sedimentary carbonate bodies were found. The carbonate rock is always associated with serpentine. The author (p. 75)

quotes Eskola (1933) with respect to the importance of the relationship of serpentine and carbonate: "This is one of the cardinal points in the great general problems of serpentine rocks associated with carbonate rocks in many ancient mountain chains." Haapla concludes that the evidence indicates a metasomatic origin rather than a sedimentary origin for the dolomitic rocks associated with these serpentines. "A more or less intense replacement of the ultramafics by carbonates" has occurred (p. 74) and, "the carbonate rocks surround, as a rule, zonally the serpentine bodies" (p. 75). Amphiboles occur in serpentine and at contacts. Serpentinization of amphiboles is a general feature, with carbonate replacing both amphiboles and serpentine. The postulated sequence of events is amphibolization, serpentinization, carbonization and talc formation. The contact amphiboles are believed to be later than the serpentinization and to be formed by hydrothermal solutions related to granitic intrusion, as is the talc and possibly the carbonate. "The areal distribution of the intensity of serpentinization is in no way related to the proximity or abundance of granite." (p. 72) In some places the degree of serpentinization increases towards the center of a body. It is concluded that the serpentine is autometamorphic.

Francis (1956) describes a serpentine body in Scotland which intrudes psammitic schist with amphibolite sills and lenticles, kyanite-garnet schist, and impure marble. This mass is part of a serpentine belt along the axis of the Scottish Highlands which is "not localized along fault lines," but is "more like the

Appalachian belt." (p. 215). The ultramafic was intruded after metamorphism and folding of the country rock. Granite intrusions came later. Small, scattered, irregular bodies, occurring at or near the serpentine margin, have actinolite, anthophyllite, chlorite, talc, and magnesite as typical minerals. There is an increase of talc towards the contacts. Prisms of actinolite occur, set in a ground mass of chlorite, talc, and magnesite. Accessories in the marginal bodies include calcite, pyrite, magnetite, hematite, ilmenite, and chromite. No zoning is found. Thin section study shows the serpentine body to have widespread relict olivine and "bastite" pseudomorphs. Francis feels that ultramafic intrusion was as a mush of crystals. The country rock schists are poor in water and there is no evidence that water came from them. The water of serpentinization is postulated to have come with the original intrusion, with later granite causing alteration of the pre-existing serpentine to talc, actinolite, chlorite, and others. Fine-grained tremolite-actinolite formed during serpentinization and later coarse-grained material formed by hydrothermal alteration. The intrusion was followed by replacement of mesh serpentine by antigorite, possibly due to high pressure, shearing stress, or thermal effects. Several powder X-ray diffraction patterns of antigorite are given and discussed.

APPENDIX B.

CHEMICAL ANALYSES OF BIOTITE AND ACTINOLITE SAMPLES.

	HAPLO GRANITE		AVERAGE	STANDARD COMPOSIT'N	G-1		AVERAGE	35 LABORATORY AVERAGE
SiO_2	72.25	72.88	72.56	72.64	72.74	72.88	72.81	72.36
TiO_2	NONE	NONE	NONE	NONE	.24	.23	.24	.25
Al_2O_3	16.13	16.40	16.26	15.78	14.40	14.41	14.40	14.44
Fe_2O_3	NONE	NONE	NONE	NONE	1.06	1.00	1.03	.93
FeO	NONE	NONE	NONE	NONE	.86	.90	.88	.99
MnO	NONE	NONE	NONE	NONE	.02	.02	.02	.03
MgO	.84	.77	.81	.80	.37	.33	.35	.39
CaO	1.79	1.73	1.76	1.82	1.32	1.30	1.31	1.41
Na_2O	3.37	3.30	3.34	3.19	3.37	3.53	3.45	3.25
K_2O	5.76	5.76	5.76	5.76	5.56	5.67	5.62	5.42
H_2O	NONE	NONE	NONE	NONE	.33	.35	.34	.37
P_2O_5	NONE	NONE	NONE	NONE	.09	.13	.11	.09
TOTAL	100.14	100.84	100.49	99.99	100.36	100.75	100.56	99.92

ANALYSES OF STANDARDS G-1 AND HAPLO GRANITE

	W-1		Average	35 Laboratry Average
SiO_2	52.92	53.42	53.17	52.34
TiO_2	1.09	1.09	1.09	1.10
Al_2O_3	14.62	14.80	14.71	15.07
Fe_2O_3	1.63	1.56	1.59	1.50
FeO	8.61	8.68	8.64	8.71
MnO	.16	.15	.16	.17
MgO	6.74	6.82	6.78	6.63
CaO	10.74	10.76	10.75	10.96
Na_2O	2.32	2.32	2.32	2.00
K_2O	.70	.72	.71	.63
H_2O	.55	.51	.53	.56
P_2O_5	.18	.21	.19	.13
TOTAL	100.26	101.04	100.64	99.79

ANALYSES OF STANDARD W-1

	152		Average	153		Average	157		Average
SiO_2	38.74	39.40	39.07	38.05	38.00	38.03	39.30	39.28	39.29
TiO_2	.10	.10	.10	.12	.13	.13	.04	.02	.03
Al_2O_3	15.89	15.81	15.85	17.58	18.05	17.81	18.21	18.39	18.30
Fe_2O_3	1.89	1.81	1.85	1.84	1.98	1.91	1.99	1.83	1.91
FeO	5.64	5.60	5.62	7.78	7.86	7.82	5.00	5.04	5.02
MnO	.05	.08	.07	.10	.10	.10	.02	.04	.03
MgO	20.29	20.05	20.17	18.15	18.09	18.09	20.75	21.10	20.93
CaO	1.54	1.60	1.57	.62	.72	.67	None	None	None
Na_2O	.20	.30	.25	.23	.21	.22	.25	.18	.22
K_2O	9.26	8.86	9.06	9.22	9.35	9.29	10.08	9.62	9.85
H_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
P_2O_5	1.45	1.49	1.47	.69	.67	.68	.11	.09	.10
Total	95.05	95.10	95.08	94.38	95.10	94.75	95.75	95.59	95.68

Biotite Analyses 152, 153, and 157

	158		Average	160		Average	167		Average
SiO_2	40.31	40.14	40.22	42.14	41.97	42.05	40.14	39.83	39.98
TiO_2	.16	.14	.15	.02	.06	.04	.76	.74	.75
Al_2O_3	15.69	16.21	15.95	14.09	14.44	14.26	15.82	16.23	16.02
Fe_2O_3	1.57	1.59	1.58	1.59	1.61	1.60	1.96	2.08	2.02
FeO	5.72	5.62	5.67	4.95	5.05	5.00	5.99	5.98	5.99
MnO	.02	.05	.04	.06	.05	.06	.02	.07	.04
MgO	21.49	20.92	21.20	22.69	21.99	22.34	21.37	21.64	21.51
CaO	None	None	None	None	None	None	None	None	None
Na_2O	.25	.30	.28	.22	.19	.21	.35	.32	.34
K_2O	9.84	10.17	10.01	9.55	9.93	9.74	9.01	8.91	8.96
H_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
P_2O_5	.14	.10	.12	.03	.07	.05	.04	.03	.04
TOTAL	95.19	95.24	95.22	95.34	95.36	95.35	95.45	95.83	95.65

BIOTITE ANALYSES 158, 160, AND 167

	172		Average	176		Average	206		Average
SiO_2	40.42	39.79	40.10	38.05	37.81	37.93	40.14	39.53	39.83
TiO_2	1.13	1.14	1.14	.05	.08	.07	.20	.19	.20
Al_2O_3	16.67	17.15	16.91	19.16	19.28	19.22	16.61	17.06	16.83
Fe_2O_3	2.25	2.43	2.34	3.07	3.00	3.04	1.72	1.89	1.80
FeO	7.96	7.96	7.96	4.02	4.11	4.07	5.39	5.28	5.34
MnO	.08	.11	.09	.04	.04	.04	.27	.27	.27
MgO	17.79	18.17	17.98	21.51	21.78	21.65	20.44	20.57	20.51
CaO	None	None	None	None	None	None	.22	.28	.25
Na_2O	.51	.42	.46	.34	.27	.31	.23	.18	.21
K_2O	9.03	9.00	9.02	9.44	9.33	9.39	9.96	9.89	9.92
H_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
P_2O_5	.04	.03	.04	.06	.05	.05	.37	.34	.36
Total	95.88	96.20	96.04	95.74	95.75	95.77	95.55	95.48	95.52

Biotite Analyses 172, 176, and 206

	208		Average	209		Average
SiO_2	40.63	40.14	40.39	40.73	40.14	40.44
TiO_2	.11	.10	.11	.12	.11	.12
Al_2O_3	18.29	18.21	18.25	16.06	16.01	16.04
Fe_2O_3	1.15	1.11	1.13	1.49	1.47	1.48
FeO	3.45	3.52	3.49	2.90	2.89	2.90
MnO	.19	.21	.20	.06	.06	.06
MgO	21.15	21.59	21.37	24.27	24.56	24.42
CaO	None	None	None	None	None	None
Na_2O	.26	.19	.23	.24	.18	.21
K_2O	10.34	10.29	10.31	9.57	9.56	9.56
H_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
P_2O_5	.13	.11	.12	.11	.08	.10
Total	95.70	95.47	95.60	95.55	95.06	95.33

Biotite Analyses 208 and 209

	210		Average
SiO_2	40.56	39.90	40.23
TiO_2	.05	.04	.05
Al_2O_3	18.65	19.14	18.90
Fe_2O_3	1.29	1.40	1.35
FeO	3.40	3.33	3.37
MnO	.08	.09	.09
MgO	21.17	21.75	21.46
CaO	None	None	None
Na_2O	.28	.28	.28
K_2O	10.24	10.06	10.15
H_2O	n.d.	n.d.	n.d.
P_2O_5	.09	.08	.09
TOTAL	95.81	96.07	95.97

210 Ledoux & Co.	
SiO_2	37.94
TiO_2	.02
Al_2O_3	20.52
TOTAL Fe	
To FeO	2.78
MnO	.12
MgO	21.54
CaO	.35
Na_2O	1.30
K_2O	10.48
Loss on Ignition	4.63
F	.25
Li_2O	.26
P_2O_5	TRACE
Boron	.005
Barium	.03
Beryllium	.002
Molybdenum	.02
TOTAL	100.25

210 – Booth, Garrett, & Blair	
SiO_2	31.46
TiO_2	.08
Al_2O_3	21.86
TOTAL Fe	
To Fe_2O_3	5.83
MnO	.07
MgO	22.15
CaO	.05
Na_2O	.53
K_2O	10.05
Loss on Ignition (1000°)	4.38
F	5.64
TOTAL	102.10

Analyses of Biotite Sample 210

	152		Average	154		Average
SiO_2	53.77	53.36	53.56	51.29	51.36	51.33
TiO_2	.02	.02	.02	.02	.01	.02
Al_2O_3	5.78	5.41	5.59	7.90	7.50	7.70
Fe_2O_3	1.78	1.61	1.70	1.68	1.81	1.74
FeO	3.76	3.98	3.87	4.01	3.80	3.90
MnO	.26	.25	.26	.18	.19	.19
MgO	19.72	19.97	19.85	19.46	19.51	19.49
CaO	12.02	12.34	12.18	11.69	12.06	11.88
Na_2O	.86	.80	.83	1.06	1.08	1.07
K_2O	.45	.44	.45	.84	.87	.86
H_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
P_2O_5	.04	.04	.04	.04	.04	.04
TOTAL	98.46	98.22	98.35	98.17	98.23	98.22

ACTINOLITE ANALYSES 152 AND 154

	159		Average	161		Average
SiO_2	56.31	55.96	56.14	54.02	53.96	53.99
TiO_2	.01	None	.01	.01	None	.01
Al_2O_3	2.07	1.84	1.95	1.35	1.25	1.30
Fe_2O_3	.73	.87	.80	.78	.73	.76
FeO	2.96	3.02	2.99	2.84	2.88	2.86
MnO	.12	.09	.11	.15	.15	.15
MgO	21.92	22.13	22.02	20.59	20.59	20.59
CaO	12.78	12.94	12.86	13.14	13.54	13.34
Na_2O	.45	.39	.42	.31	.25	.28
K_2O	.23	.27	.25	.24	.20	.22
H_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
P_2O_5	Trace	.02	.01	None	None	None
Total	97.58	97.53	97.56	93.43	93.55	93.50

Actinolite Analyses 159 and 161

	166		Average	171		Average	175		Average
SiO_2	57.09	56.52	56.80	56.57	56.07	56.32	58.66	58.03	58.34
TiO_2	.02	.01	.02	.01	None	.01	.01	None	.01
Al_2O_3	1.35	1.27	1.31	1.82	1.75	1.78	.03	None	.02
Fe_2O_3	.95	.82	.89	.92	1.04	.98	.59	.53	.56
FeO	3.42	3.55	3.48	3.82	3.78	3.80	2.23	2.34	2.28
MnO	.17	.14	.16	.18	.16	.17	.12	.08	.10
MgO	22.04	21.90	21.97	21.77	21.93	21.85	24.73	25.06	24.90
CaO	12.03	12.42	12.22	11.78	11.98	11.88	9.85	10.22	10.04
Na_2O	.36	.36	.36	.44	.43	.44	.19	.19	.19
K_2O	.18	.22	.20	.23	.26	.25	.06	.09	.08
H_2O	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
P_2O_5	.11	.09	.10	.02	Trace	.01	Trace	None	Trace
TOTAL	97.72	97.30	97.51	97.56	97.40	97.49	96.47	96.54	96.52

Actinolite Analyses 166, 171, and 175

	BIOTITE	BIOTITE
F	.76	
SiO_2	39.08	39.55
TiO_2		
Al_2O_3	15.38	15.95
Fe_2O_3		
FeO	7.12	7.80
MnO	.31	
MgO	23.58	22.25
CaO		
Na_2O	2.63	10.35
K_2O	7.50	
H_2O-	2.24	4.10
H_2O+		
P_2O_5		
TOTAL	98.60	100.00
LOCATION	CHESTER, MASS.	CHESTER, MASS.
TYPE OF CONTACT	AT CONTACT OF PLAGIOCLASE VEIN IN TALC-CHLORITE SEAM	
REFERENCE	EMERSON 1895	EMERSON 1895

	BIOTITE	BIOTITE
F	4.34	3.17
SiO_2	42.02	39.97
TiO_2	1.35	2.64
Al_2O_3	18.75	17.51
Fe_2O_3	.66	2.26
FeO	8.29	14.81
MnO	.27	.22
MgO	9.55	8.45
CaO	.93	NONE
Na_2O	.73	.45
K_2O	8.54	8.48
H_2O-	.16	.32
H_2O+	2.44	2.48
Li_2O	1.20	.65
Rb_2O	1.85	1.48
Cs_2O	.47	1.12
TOTAL	101.55	104.01
LOCATION	KINGS MT., N.C.	TIN MT., S.D.
TYPE OF CONTACT	MICA SCHIST-PEGMATITE CONTACT	MICA SCHIST-PEGMATITE CONTACT
REFERENCE	HESS & STEVENS 1937	HESS & STEVENS 1937

PUBLISHED ANALYSES OF CONTACT BIOTITE AND ACTINOLITE SAMPLES

	Biotite	Biotite	Biotite
F	2.89	1.67	Present but n.d.
SiO_2	40.80	37.39	38.72
TiO_2	.05	2.48	.47
Al_2O_3	13.16	20.17	18.39
Fe_2O_3	1.84	2.08	1.05
FeO	2.16	14.32	9.54
MnO		n.d.	.16
MgO	26.72	9.80	18.44
CaO	.46	.51	Trace
Na_2O	2.44	.87	.68
K_2O	7.95	8.56	9.74
H_2O-	~~2.95~~	~~3.03~~	.16
H_2O+			.53
P_2O_5			.11
Cr_2O_3		.02	
Total	101.42	100.88	97.99
Location	Mansjö, Sweden	Dutchess County, New York	Chester, Vt.
Type of Contact	Pegmatite – Limestone Contact	Pegmatite – Schist Contact	Serpentine – Schist Contact
Reference	v. Eckermann 1925	Barth 1936	Phillips & Hess 1936

	Actinolite
F	
SiO_2	53.27
TiO_2	.06
Al_2O_3	4.92
Fe_2O_3	1.43
FeO	6.78
MnO	.20
MgO	19.47
CaO	11.70
Na_2O	.75
K_2O	.59
H_2O-	.05
H_2O+	.39
P_2O_5	.11
Total	99.72
Location	Chester, Vt.
Type of Contact	Serpentine – Schist Contact
Reference	Phillips & Hess 1936

Published Analyses of Contact Biotite and Actinolite Samples

APPENDIX C.

CALCULATION OF CHEMICAL ANALYSES TO AN ATOMIC BASIS.

BIOTITE BASIS: 12 (O, OH) ATOMS

ACTINOLITE BASIS: 24(O, OH) ATOMS

Sample	Bio. #152	Bio. #153	Bio. #157	Bio. #158	Bio. #160	Bio. #167	Bio. #172	Bio. #176
Atoms								
SiO_2	2.758	2.704	2.780	2.846	2.955	2.836	2.871	2.684
TiO_2	.005	.007	.002	.008	.002	.040	.061	.004
Al_2O_3	1.318	1.492	1.526	1.330	1.181	1.345	1.427	1.603
Fe_2O_3	.098	.102	.102	.084	.084	.108	.126	.162
FeO	.332	.465	.297	.335	.294	.355	.475	.240
MnO	.004	.006	.002	.002	.004	.002	.005	.002
MgO	2.121	1.916	2.206	2.235	2.338	2.273	1.917	2.282
CaO	.119	.051	none	none	none	none	none	none
Na_2O	.034	.030	.030	.038	.029	.047	.064	.043
K_2O	.815	.842	.888	.904	.874	.810	.823	.847
P_2O_5	.088	.041	.006	.007	.003	.002	.002	.003
H_2O	2.316	2.489	2.038	2.255	2.178	2.057	1.890	1.996

Sample	Bio. #206	Bio. #208	Bio. #209	Bio. #210	Biotite (Phillips & Hess - 1936)	Actinolite (Phillips & Hess - 1936)
Atoms						
SiO_2	2.818	2.831	2.821	2.827	2.843	7.619
TiO_2	.011	.006	.006	.003	.026	.006
Al_2O_3	1.403	1.507	1.319	1.565	1.591	.829
Fe_2O_3	.096	.060	.078	.071	.058	.154
FeO	.316	.204	.169	.198	.585	.811
MnO	.016	.012	.004	.005	.010	.024
MgO	2.163	2.231	2.538	2.246	2.017	4.148
CaO	.019	none	none	none	trace	1.792
Na_2O	.029	.031	.028	.038	.097	.208
K_2O	.895	.922	.850	.909	.912	.108
P_2O_5	.022	.007	.006	.005	.007	.013
H_2O	2.114	2.056	2.172	2.246	1.322	.687

Samples	Act. #152	Act. #154	Act. #159	Act. #161	Act. #166	Act. #171	Act. #175
Atoms							
SiO_2	7.571	7.205	7.712	7.173	7.802	7.755	7.829
TiO_2	.002	.002	.001	.001	.002	.001	.001
Al_2O_3	.930	1.273	.316	.204	.212	.288	.003
Fe_2O_3	.181	.184	.083	.076	.092	.101	.057
FeO	.457	.457	.344	.318	.399	.437	.256
MnO	.031	.023	.013	.017	.019	.020	.011
MgO	4.180	4.075	4.506	4.075	4.495	4.482	4.977
CaO	1.844	1.786	1.892	1.898	1.798	1.752	1.443
Na_2O	.227	.291	.112	.072	.096	.117	.049
K_2O	.081	.154	.044	.037	.035	.044	.013
P_2O_5	.005	.005	.001	none	.012	.001	none
H_2O	1.555	1.666	2.235	5.758	2.280	2.304	3.114

Sample	Calculated Atomic Formula
Bio. #152	$(K,Na,Ca)_{.97}(Mg,Fe'',Fe''',Ti,Mn,Al)_{2.72}(Si,Al,P)_{4.00}O_{9.68}(OH)_{2.32}$
Bio. #153	$(\_\_\_\_\_)_{.92}(\_\_\_\_\_)_{2.73}(\_\_\_\_\_)_{4.00}O_{9.51}(OH)_{2.49}$
Bio. #157	$(K,Na)_{.92}(Mg,Fe'',Fe''',Ti,Mn,Al)_{2.92}(Si,Al,P)_{4.00}O_{9.96}(OH)_{2.04}$
Bio. #158	$(\_\_\_\_)_{.94}(\_\_\_\_\_)_{2.85}(\_\_\_\_\_)_{4.00}O_{9.74}(OH)_{2.26}$
Bio. #160	$(\_\_\_\_)_{.90}(\_\_\_\_\_)_{2.86}(\_\_\_\_\_)_{4.00}O_{9.82}(OH)_{2.18}$
Bio. #167	$(\_\_\_\_)_{.86}(\_\_\_\_\_)_{2.96}(\_\_\_\_\_)_{4.00}O_{9.94}(OH)_{2.06}$
Bio. #172	$(\_\_\_\_)_{.89}(\_\_\_\_\_)_{2.88}(\_\_\_\_\_)_{4.00}O_{10.11}(OH)_{1.89}$
Bio. #176	$(\_\_\_\_)_{.89}(\_\_\_\_\_)_{2.98}(\_\_\_\_\_)_{4.00}O_{10.00}(OH)_{2.00}$
Bio. #206	$(\_\_\_\_,Ca)_{.92}(\_\_\_\_\_)_{2.85}(\_\_\_\_\_)_{4.00}O_{9.89}(OH)_{2.11}$
Bio. #208	$(\_\_\_\_)_{.95}(\_\_\_\_\_)_{2.86}(\_\_\_\_\_)_{4.00}O_{9.94}(OH)_{2.06}$
Bio. #209	$(\_\_\_\_)_{.88}(\_\_\_\_\_)_{2.94}(\_\_\_\_\_)_{4.00}O_{9.83}(OH)_{2.17}$
Bio. #210	$(\_\_\_\_)_{.95}(\_\_\_\_\_)_{2.92}(\_\_\_\_\_)_{4.00}O_{10.11}(OH)_{1.89}$
Biotite (Phillips & Hess - 1936)	$(\_\_\_\_)_{1.01}(\_\_\_\_\_)_{3.14}(\_\_\_\_\_)_{4.00}O_{10.68}(OH)_{1.32}$
Act. #152	$(Ca,Na,K)_{2.15}(Mg,Fe'',Fe''',Ti,Mn,Al)_{5.36}(Si,Al,P)_{8.00}O_{22.44}(OH)_{1.56}$
Act. #154	$(\_\_\_\_\_)_{2.23}(\_\_\_\_\_)_{5.22}(\_\_\_\_\_)_{8.00}O_{22.33}(OH)_{1.67}$
Act. #159	$(\_\_\_\_\_)_{2.05}(\_\_\_\_\_)_{4.98}(\_\_\_\_\_)_{8.00}O_{21.76}(OH)_{2.24}$
Act. #161	$(\_\_\_\_\_)_{2.01}(\_\_\_\_\_)_{4.49}(\_\_\_\_\_)_{7.38}O_{18.24}(OH)_{5.76}$
Act. #166	$(\_\_\_\_\_)_{1.93}(\_\_\_\_\_)_{5.03}(\_\_\_\_\_)_{8.00}O_{21.72}(OH)_{2.28}$
Act. #171	$(\_\_\_\_\_)_{1.91}(\_\_\_\_\_)_{5.09}(\_\_\_\_\_)_{8.00}O_{21.70}(OH)_{2.30}$
Act. #175	$(\_\_\_\_\_)_{1.51}(\_\_\_\_\_)_{5.30}(\_\_\_\_\_)_{7.83}O_{20.89}(OH)_{3.11}$
Actinolite (Phillips & Hess - 1936)	$(\_\_\_\_\_)_{2.11}(\_\_\_\_\_)_{5.60}(\_\_\_\_\_)_{8.00}O_{23.31}(OH)_{.69}$

BIBLIOGRAPHY

Atwater, W. (1821), Hill of Serpentine: Am. Jour. Sci., 1st ser., v. 3, p. 238-239.

Barth, T. F. W. (1936), Structural and Petrologic Studies in Dutchess County, New York: Geol. Soc. America Bull., v. 47, p. 775-850.

Benson, W. N. (1918), The Origin of Serpentine; A Historical and Comparative Study: Am. Jour. Sci., 4th Ser., v. 46, p. 693-731.

_____ (1926), The Tectonic Conditions Accompanying the Intrusion of Basic and Ultrabasic Igneous Rocks: U. S. Nat. Acad. Sci., Mem. 1, v. 19.

Bowen, N. L. (1928), The Evolution of the Igneous Rocks: Princeton Univ. Press, Princeton, N. J., p. 167.

_____, and Schairer, J. F. (1936), The Problem of the Intrusion of Dunite in the Light of the Olivine Diagram: 16th Internat. Geol. Congr. Repts. (1933), p. 391-396.

_____, and Tuttle, O. F. (1949), The System MgO-SiO_2-H_2O: Geol. Soc. America Bull., v. 60, p. 439-460.

Cairnes, C. E. (1930), The Serpentine Belt of Coquihalla Region, Yale District, British Columbia: Geol. Surv. Canada, Summ. Rept., 1929, pt. A.

Chidester, A. H. et al. (1951), Talc Investigations in Vermont: U.S. Geol. Surv., Circular 95.

Crosby, W, O. (1896), Geological Report on Verd Antique Marble Deposit: Clark & Story, Westfield, Massachusetts.

Davis. E. (1826), Notice of Rocks and Minerals in Westfield, Massachusetts: Am. Jour. Sci., 1st ser., v. 10, p. 213-217.

Dennen, W. H. (1951), Variations in Chemical Composition Across Igneous Contacts: Geol. Soc. America Bull., v. 62, p. 547-558.

DuRietz, T. (1935), Peridotites, Serpentines, and Soapstones of Northern Sweden: Geol. Fören. Förh., v. 57, p. 133-260.

Eaton, A. (1819), Account of the Strata Perforated by, and of the Minerals Found in, the Great Adit to the Southampton Lead Mine: Am. Jour. Sci., 1st ser., v. 1, p. 136-139.

Eckermann, H. V. (1925), The Glimmermineralien im Mansjo-Kalk-kontakt: Tschmer. Mineral. Petr. Mitt., v. 38. p. 277-287.

Emerson, B. K. (1895), A Mineralogical Lexicon of Franklin, Hampshire, and Hampden Counties, Massachusetts: U. S. Geol. Surv., Bull. 126.

_____ (1898), Geology of Old Hampshire County, Massachusetts: U. S. Geol. Surv., Mon. 29.

_____ (1917), Geology of Massachusetts and Rhode Island: U. S. Geol. Surv., Bull. 597.

Eskola, P. (1933), On the Chrome Minerals of Outokumpu: Soc. Géol. Finlande, C. R., v. 7, Comm. Géol. Finlande, Bull., v. 103, p. 26.

Fairbairn, H. W. (1953), Precision and Accuracy of Chemical Analysis of Silicate Rocks: Geochim. Cosmochim. Acta, v. 4, p. 143-156.

_____ et al. (1951), A Cooperative Investigation of Precision and Accuracy in Chemical, Spectrochemical and Model Analysis of Silicate Rocks: U. S. Geol. Surv., Bull. 980, 71.p.

_____, and Schairer, J. F. (1952), A Test of The Accuracy of Chemical Analysis of Silicate Rocks: Am. Mineral., v. 37, p. 744-757.

Francis, G. H. (1956), The Serpentinite Mass in Glen Urquhart, Inverness-shire, Scotland: Am. Jour. Sci., v. 254, p. 201-226.

Fyfe, W. S., Turner, F. J., and Verhoogen, J. (1958), Metamorphic Reactions and Metamorphic Facies: Geol. Soc. America, Mem. 73, 259 p.

Gillson, J. L. (1927), Origin of the Vermont Talc Deposits: Econ. Geol., v. 22, p. 246-287.

Groves, A. W. (1951), Silicate Analysis: George Allen & Unwin, Ltd., London, p. 95-104.

Haapla, P. (1936), On the Serpentine Rocks in Northern Karelia: Comm. Geol. Finlande, Bull., v. 114.

Hall, A. J. (1941), The Relation Between Color and Chemical Composition in the Biotites: Am. Mineral., v. 26, No. 1, p. 29-41.

Henrich, E. W. (1946), Studies in the Mica Group; The Biotite-Phlogopite Series: Am. Jour. Sci., v. 244, p. 836-848.

_____ et al. (1953), Studies in the Natural History of Micas: Univ. of Michigan Engineering Research Inst.

Hess, H. H. (1933), The Problem of Serpentinization and the Origin of Certain Crysotile Asbestos, Talc and Soapstone Deposits: Econ. Geol., v. 28, p. 634-657.

_____(1937), Geological Interpretation of Data Collected on Cruise of USS Baracuda in the West Indies: Am. Geophys. Union, Trans., 18th Annual Meeting, p. 69-77.

_____ (1938), A Primary Peridotite Magma: Am. Jour. Sci., 5th ser., v. 35, p. 321-344.

_____ (1939), Island Arcs, Gravity Anomalies and Serpentinite Intrusions: Internat. Geol. Congr., Moscow, 1937, Rept. 17, v. 2, p. 263-283.

_____ (1955), Serpentines, Orogeny, and Epeirogeny: Geol. Soc. America, Spec. Paper 62, p. 391-407.

Hess, F. L., and Stevens, R. E. (1937), Am. Mineral., v. 22, p. 1040.

Hitchcock, E. (1841), Final Report of the Geology of Massachusetts: J. S. & C. Adams, Amherst, Mass., v. 2, p. 617-618.

Hunt, T. S. (1854), On Some of the Crystalline Limestones of North America: Am. Jour. Sci., 2nd ser., v. 18, p. 193-200.

Korzhinsky (Korjinsky), D. S. (1936), Mobility and Inertness of Components in Metasomatosis: Acad. Sci., U.S.S.R., Bull., Sér. Géol., 1, p. 35-60.

Kranck, S. H. (1959), Chemical Petrology of Metamorphic Iron Formations and Associated Rocks in the Mount Reed Area in Northern Quebec, Ph.D. Thesis, Massachusetts Institute of Technology.

Macdonald, G. A. (1941), Progressive Metasomatism of Serpentine in the Sierra Nevada of California: Am. Mineral., v. 26, p. 276-287.

Martin, G. C. (1898), An Occurrence of Dunite in Western Massachusetts: Am. Jour. Sci., 4th ser., v. 6, p. 244-248.

Mercey, E. L. P. (1956), The Accuracy and Precision of "Rapid Methods" of Silicate Analysis: Geochim. Cosmochim. Acta, v. 9, p. 161-173.

Nockolds, S. R. (1947), The Relation Between Chemical Composition and Paragenesis in the Biotite Micas of Igneous Rocks: Am. Jour. Sci., v. 245, p. 401-420.

Otis, L. D. (1952), Minerals and Rocks of Springfield and Vicinity: Mus. Nat. Hist., Springfield, Massachusetts, Bull. 9, 50 p.

Pabst, A. (1942), The Mineralogy of Metamorphosed Serpentine at Humphreys, Fresno County, California: Am. Mineral., v. 27, p. 570-585.

Palache (1907), Occurrence of Olivine in the Serpentine of Chester and Middlefield, Massachusetts: Am. Jour. Sci., 4th ser., v. 24, p. 491-495.

Phillips, A.H., and Hess, H. H. (1936), Metamorphic Differentiation at Contacts between Serpentinite and Siliceous Country Rocks: Am. Mineral., v. 21, p. 333-362.

Phinney, W. C. (1959), Phase Equilibrium in the Metamorphic Rocks of St. Paul Island and Cape North, Nova Scotia, Ph.D. Thesis, Massachusetts Institute of Technology.

Pitcher, W. S., and Sinha, R. C. (1958), The Petrochemistry of the Andara Aureole: Geol. Soc. London, Quart. Jour., v. 113, p. 393-408.

Pratt, J. H., and Lewis, J. V. (1905), Corundum and the Peridotites of Western North Carolina: Geol. Surv. North Carolina, Rept., v. 1, 464 p.

Read, H. H. (1934), On Zoned Associations of Antigorite, Talc, Actinolite, Chlorite, and Biotite in Unst, Shetland Islands: Mineral. Mag., v. 23, p. 519-540.

Riley, J. P. (1958a), The Rapid Analysis of Silicate Rocks and Minerals: Anal. Chim. Acta, v. 19, p. 413-428.

_____ (1958b), Simultaneous Determination of Water and Carbon Dioxide in Rocks and Minerals: Analyst, v. 83, p. 42-49.

Shapiro, L., and Brannock, W. W. (1956), Rapid Analysis of Silicate Rocks; A Contribution to Geochemistry: U. S. Geol. Surv., Bull. 1036-C, 56 p. (revised from Circular 165).

Steinmann, G. (1905), Geologische Beobachtungen in den Alpen. II: Die Schardt'sche Uberfaltungstheorie und die Geologische Bedeutung der Tiefseeabsätze und der Ophiolithischen Massengesteine: Ber. Nat. Ges. Freiburg, Bd. 16, p. 44-65.

Taliaferro, N. L. (1943), Franciscan Knoxville Problem: Am. Assoc. Pet. Geol., Bull., v. 27, p. 109-219.

Thompson, J. B., Jr. (1959), Equilibrium in Metasomatic Processes: Researches in Geochemistry, P. H. Abelson, Editor, John Wiley & Sons, Inc., N. Y., N. Y., 511 p.

Turner, F. J., and Verhoogen, J. (1951), Igneous and Metamorphic Petrology: McGraw-Hill Book Co., Inc., New York, p. 423.

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